

# Recent progress in sodium alginate based sustainable hydrogels for environmental applications

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## ABSTRACT

Recently, there is a growing research interest in the applications and development of novel sustainable hydrogel materials in waste water treatment because of radically distinctive chemical and physical characteristics of hydrogels such as hydrophilicity, swell ability and modifiability to name a few. Hydrogels have exposed the hypernym functioning in the removal of a wide range of aqueous pollutants containing toxic dyes and heavy metal ions. A large amount of water gets incorporated in the three dimensional networks of hydrophilic structures of hydrogels. The prime objective of this review article is to render a presentation on the recent advances in the modifications of sodium alginate based hydrogels for the adsorptive removal of toxic pollutants. In addition, article also briefly gives the classification and properties of hydrogels and alginate.

**Keywords:** Alginate; sodium alginate based hydrogel; dye and metal removal; water purification.

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## 10 **1. Introduction**

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12 Recently, there has been a noticeable growth in the development of hydrogels composite and  
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14 their applications in various fields including environmental remediation (Khan and Lo, 2016a; Li  
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16 et al., 2013; Sahiner and Seven, 2014; Thakur et al., 2015, 2017b, 2017a). In regard to  
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18 wastewater treatment, hydrogels based composites have exhibited superior performance in the  
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20 adsorptive removal of different types of inorganic pollutants as well as organic pollutants  
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22 (Mahdavinia et al., 2016; Ngah and Fatinathan, 2008; rights are reserved by Shivani and Shetye,  
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24 n.d.). In many studies, hydrogels composites have clearly outcompeted the conventional  
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26 adsorbents especially in terms of adsorption capacity (Burkert et al., 2007). Hydrogels are three-  
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28 dimensional, polymeric networks capable of absorbing large amount of water (Zhao et al., 2013).  
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30 Hydrogels based composites have been tailored to grab most of the applications to get more  
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32 functional features such as maximum equilibrium swelling, high rate of absorption, less residual  
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34 content, porosity, uniformly soft, high durability, odorless, photo-stability (Ahmed, 2015; Gao et  
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36 al., 2011). Hydrogels composites are chemically and physically stable, smooth, flexible  
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38 polymeric network, reusable and multi-functional in nature. These flexible polymeric materials  
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40 upholds the ability to swell and retain a consequential amount of water within their structure, but  
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42 without being dissolved in water (Shetye et al., 2015). In the past 50 years, hydrogels have  
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44 received appreciable attention from worldwide research community, due to their wide-ranging  
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46 research interest moreover, many application holdings make this field more acceptable (Khan  
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48 and Lo, 2016a). Research interest in the field of environmental science has been increasing with  
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50 emphasis on use of bio-based hydrogels for advanced applications (Khan and Lo, 2016a). One of  
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4 the major challenges is the recovery of hydrogel from solution after the completion of the  
5 adsorption or desorption process, especially in micro or nano-sized hydrogel (Izawa and  
6 Kadokawa, 2010). The selective removal and recovery of a targeted pollutants are the challenges  
7 because the co-present ingredients in wastewater often create unwanted competition and  
8 interference (F. Zhao et al., 2015). Furthermore, there have been some stability concerns for  
9 hydrogels composites which affect their fate and reusability (Roy et al., 2010).

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19 It's been about many decades since research work is going on hydrogels and this field may come  
20 up with the dream of drought-free planet by purifying waste water (Fane et al., 2015). Although  
21 water purification by hydrogels is well set up by the various adsorption techniques but it quietly  
22 necessitates further improvements concerning degradability, intrinsic cellular interactions and  
23 strong mechanical properties in the different dynamic environment (Du et al., 2015). After  
24 acknowledging different research works of this field, we can define the ideal characteristics of  
25 hydrogels, along with the novel advancements in the material science (Vermonden et al., 2012a).  
26 Moreover, by overcoming the limitations of hydrogels in water treatment domain, we may build  
27 faith of society on water purification technologies because it promises the most practical and  
28 economic option for water purification from industrial and laboratory waste. In this article, we  
29 have made a platform for the upgraded hydrogel networks in relevance to the water treatment  
30 mission. In addition, template hydrogel materials stands up with many benefits in different fields  
31 like chemical catalysis, nano-engineering, nano-medicines, nano-energy and especially for  
32 reactions occurring in aqueous media (**Figure 1**) (Sahiner, 2013). The proven efficiencies of  
33 different chemical modifications has now been attracting the researchers to redesign and  
34 synthesize hydrogels with metal nano-particles *in situ* or *ex situ* (Lee and Mooney, 2001). Most  
35 of the work that has been done in one dimensional or two dimensional materials like surfactants  
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4 or film, here is the applicable approach to generate three dimensional material with exterior and  
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6 interior porous topography offers multifunctional utility. Recently, *in situ* approach of metal  
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8 nano-particles with hydrogels is opening a new era on account of its fabulous properties such as  
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10 optical, chemical, electronic and magnetic properties (Sahiner, 2013). Although many alternates  
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12 may come out in future to solve the problem of aggregation of metal nano-particles and also their  
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14 non-symmetrical behaviour in-between flexible hydrogel network. Despite the unique  
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16 characteristics of these polymeric networks, science is yet not able to predict the quantitative  
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18 mechanical response of the hydrogels based on their physico-chemical properties which limits  
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20 our research ability to design something new (Wang et al., 2017). This review article focuses on  
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22 the recent developments in sodium alginate based hydrogels and composites for water  
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24 purification. The classification of hydrogels, synthesis of hydrogels and composites along with  
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26 the properties of alginate have been discussed in this article.  
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## 33 **2. Method**

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36 Potential adsorption from bio-renewable resources involves development of the most commercial  
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38 polymers based products for environmental application. The bio-based polymeric hydrogels have  
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40 proved their efficiencies many times and also have added an advantage of bio-degradation in an  
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42 aqueous media. Among numerous hydrogels, sodium alginate based hydrogels have allured the  
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44 research interest for number of applications due to their inbuilt physico-chemical nature. But this  
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46 research field is still in an early stage of development with bounded information regarding  
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48 literature of sodium alginate based hydrogels for water remediation. In this article, the data  
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50 collection involved several steps, started from the classification, preparation, utilization and  
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52 reusability of hydrogels to the recent advancements in sodium alginate loaded hydrogels for the  
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54 removal of organic and inorganic pollutants. The development of “Sodium Alginate based  
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4 Hydrogels” initiatives, require inclusive knowledge on alginate based materials and different  
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6 sustainable materials for new tailored properties and applications. The synthesis and applications  
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8 of novel innovative materials from alginate will improve the horizon of sustainable hydrogels as  
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10 well as possibly address the problem of lack of human friendly/biocompatible tissues and  
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12 scaffolds materials. Therefore, we summarize various efforts of researchers all around the globe  
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14 to develop new sustainable hydrogel materials for environmental applications from alginate  
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16 based polymers in the present manuscript. This perspective article in tends to show that  
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18 developed hydrogels from bio-renewable resources can be efficiently prepared and executed as  
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20 potential environmental friendly, biodegradable and economic materials. This reviews some  
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22 important questions about the potential use of “Sustainable Biomaterials” in advanced  
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24 applications including environmental engineering.  
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### 31 32 **3. Classification of Hydrogels**

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34 Hydrogel products can be classified on several bases (Ahmed, 2015):  
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#### 36 37 **3.1. Based onSource**

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39 Source means a thing or person from which something is obtained. So, on the basis of source,  
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41 these polymeric materials can be classified into two categories: synthetic or natural source  
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43 (Nayak et al., 2010). The natural polymeric materials show profitable bio-compatibility, bio-  
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45 degradability but these are far behind in case of mechanical properties. However, on the other  
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47 hand, there is a lack of bio-active properties in synthetic polymeric material.  
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#### 51 52 **3.2. According to Polymeric Composition**

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54 Composition simply means “act of combining of elements to form a whole”. On the basis of this,  
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56 hydrogel products have been categorized into the following: (a) hydrogels belong to the polymer  
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58 network consisting of basic structural monomer unit formed via homo-polymerization (Qiu and  
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4 Park, 2001). Homo-polymer hydrogels are those polymer systems which emerge from single unit  
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6 of monomers and comprises of normal auxiliary segment of any polymer framework. There  
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8 might be some possibility of having cross-linked skeletal structures, which depends upon the  
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10 polymerization technique as well as nature of the monomer being used; **(b)** co-polymeric  
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12 hydrogels are the chemical compounds of high molecular weight made up of two or more  
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14 different monomer subunits with not less than one hydrophilic component organized in a  
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16 random, block or alternating configuration across the chain of the polymer network (Thakur,  
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18 2016); **(c)** multi-polymer interpenetrating polymeric hydrogels, the salient class of hydrogels that  
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20 consist of two different cross-linked components of synthetic or natural polymer, arranged in a  
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22 network form (Shetye et al., 2015). This class is further split up into semi-interpenetrating  
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24 polymeric hydrogel in which one of the hydrogel component is non-cross-linked another is  
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26 cross-linked polymer or monomer.  
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### 33 **3.3. Based on Configuration**

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35 These are generally based on the physical arrangement and reactant (chemical) composition of  
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37 the hydrogel and can be classified as: **(a)** amorphous (no apparent organization), **(b)** semi-  
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39 crystalline (imperfectly crystalline) (Thoniyot et al., 2015a); it is a mixture of distinct as well as  
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41 indistinct (amorphous) phases, **(c)** crystalline (regular arrangement of atoms in space lattice), **(d)**  
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43 hydrogen bonded hydrogels.  
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### 48 **3.4. Based on Type of Cross-linking**

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50 This class has two different division of cross-linked network of hydrogels based on their  
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52 chemical /physical nature (Caló and Khutoryanskiy, 2015). Physical networks have been found  
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54 to exhibit temporary junctions that appear from physical relations like ionic interactions,  
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56 hydrogen bonds, or hydrophobic interactions (L. Z. Zhao et al., 2015) whereas, chemically cross-  
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4 linked networks have unchanging junctions. This is generally achieved by using physical  
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6 activities like coalition, conglomeration, connection, crystallization, difficulty and hydrogen  
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8 bonding. Conversely, a chemical procedure is a process in which incorporated bivalent cross-  
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10 linking is utilized to assemble a synthetic hydrogel. Physical hydrogels are  
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12 convertible/reversible in nature due to the informational augmentations, while synthetic  
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14 hydrogels are unchangeable/long lasting in nature. Cross-linking makes the network viscoelastic  
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16 which provides hardness and stickiness to the structure.  
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### 21 **3.5. Based on Physical Appearance**

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23 The outward composition of hydrogel film or microsphere depends on the technique of  
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25 polymerization, cross-linking or any other preparation process.  
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### 28 **3.6. Classification According to Network Electrical Charge**

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30 This classification is divided into four main types based on the presence/absence of electrical  
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32 charge pointed on the double cross-linked chains (Ahmed, 2015): **(a)** neutral (non-ionic) gels; **(b)**  
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34 ionic gels (including anionic or cationic); **(c)** amphoteric electrolyte (containing both acidic as  
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36 well as basic groups); **(d)** zwitter-ionic (having anionic and cationic groups together in each  
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38 structural repeating unit).  
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## 43 **4. Preparation of Hydrogels**

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45 Hydrogels are commonly defined as semi-solid, three-dimensional, polymeric networks obtained  
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47 from a class of synthetic or natural polymers (collagen, gelatin, starch, alginate etc.) which can  
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49 retain significant amount of water and biological fluids (Shi et al., 2011). During the hydration  
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51 process, the structure of polymeric network is created by the domains or hydrophilic groups  
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53 (Talaat et al., 2008). Preparation of hydrogels is based on hydrophilic monomers (polar part first  
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55 to be hydrated) and hydrophobic monomers (forms hydrophobically-bounded water) are  
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4 sometimes used to control the properties for specific applications (Can et al., 2007), (Thoniyot et  
5 al., 2015a). Traditionally, chemical polymerization techniques are used in case of synthetic  
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14 (Mehrali et al., 2017). Hydrogels are mainly synthesized using polar monomers and their  
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sometimes used to control the properties for specific applications (Can et al., 2007), (Thoniyot et al., 2015a). Traditionally, chemical polymerization techniques are used in case of synthetic polymers that form hydrogels. The mechanical strength of the synthetic polymers has been reported to cause slow degradation rate, but on the other hand, it provides the durability as well (Mehrali et al., 2017). Hydrogels are mainly synthesized using polar monomers and their preparation starts from initiation reaction and ends up at termination. In addition, three fundamental parts (**Figure 2**) of the hydrogels preparation are monomer, cross-linker and initiator (Ahmed, 2015). For example, Thakur et al., prepared sodium alginate based hydrogels by a polymerization technique employing acrylic acid as monomer, N,N'-methylenebisacrylamide and potassium persulphate as cross-linker and initiator respectively (Thakur and Arotiba, n.d.). Diluents like water and biological fluids can be used to control the final hydrogel properties and the heat of polymerization (Ma et al., 2010). Starting materials can be optional and is divided into synthetic polymers, natural polymers or combination of both. A number of polymerization methods can be adopted to form hydrogels: **(a)** Bulk polymerization means two or more monomers are combined with the help of suitable initiator for the formation of hydrogel. In this polymerization, monomers make homogeneous composition of hydrogels. **(b)** Solution polymerization/cross-linking means ionic and neutral monomers are reacted together with suitable cross-linking agent. The reaction is initiated with the help of UV-irradiation or using a redox initiator system. **(c)** Suspension polymerization or inverse-suspension polymerization means monomers as well as initiator are diffused as a homogenous mixture in the hydrocarbon phase. **(d)** Polymerization by irradiation means high energy radiations such as like electron beams/gamma rays etc are employed as an initiator to prepare the hydrogel (Karadağ et



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4 al., 2001) and (e) grafting to a support means for increases the mechanical properties of the  
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6 hydrogel, hydrogels are coated with the strong grafting agents.  
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## 9 **5. Preparation of Hydrogels Composite**

10 Hydrogels composite have been advanced by adding a wide range of nano-materials (metallic,  
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12 polymeric, carbon-based) (Merino et al., 2015; Shin et al., 2011; Zuo et al., 2015). Nano-  
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14 materials can be incorporated within the bulk hydrogel framework. Five basic methods have  
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16 been used to obtain a uniform distribution (Thoniyot et al., 2015a): (a) preparation of hydrogel  
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18 in a nano-particle suspension, (b) addition of the nano-particles into hydrogel matrix after  
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20 gelation, (c) reactive nano-particle preparation within a preformed hydrogel, (d) hydrogels nano-  
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22 composite via cross-linking and (e) development of hydrogel nano-composite using polymers,  
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24 metallic nano-particles and different gelator molecules (Khan and Lo, 2016a).  
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### 31 **5.1. Preparation of Hydrogel in Nano-particle Suspension**

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33 The perfect technique to build hydrogel composites is the gelation of nano-particles suspension  
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35 in hydrogel matrix. Through this technique, hydrogel composites becomes optically and  
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37 mechanically responsive (Omidian et al., 2006). In this technique, monomers, cross-linkers and  
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39 nano-particles are combined to form hydrogel composites through gelation (Thakur et al., 2016).  
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41 This technique is the easiest technique for the synthesis of hydrogel composites. Sershen et al.,  
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43 synthesized optomechanically N-isopropyl acrylamide/acrylamide hydrogel matrix by adding  
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45 gold nano-particles through gelation using initiator ammonium persulfate and accelerator  
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47 tetramethylethylenediamine (Sershen et al., 2002). The hydrogel applications in the mechanical  
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49 field can be enhanced by adding the nano-particles into the hydrogel matrix.  
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### 55 **5.2. Addition of Nano-particles into Hydrogel Matrix after Gelation**

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4 In this method, nano-particles are added in the hydrogel matrix after the gelation process. Yissar  
5 et al., added gold nano-particles into polymer hydrogel matrix after the electro polymerization  
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7 (Pardo-Yissar et al., 2001). Electro polymerization cannot be achieved in the presence of the  
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9 gold nano-particles because of aggregation of gold nano-particles in electric field (Kim and  
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11 Randall Lee, 2006). In this work, cross-linked polyacrylamide gels were prepared on gold-wire  
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13 electrodes by the electro-polymerization of acrylamide along with  $ZnCl_2$  and N, N'  
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15 methylenebisacrylamide.  
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### 21 **5.3. Reactive Nano-particle Preparation within Preformed Hydrogel**

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23 Langer's group formulated this technique, which consists of precursors loading into a hydrogel  
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25 matrix, rather than formed nano-particles (Wang et al., 2004). For example, hydrogel network  
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27 containing embedded Au (III) ions with thiol groups was produced. Hydrogel matrix containing  
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29 thiol group was able to modulate gold nano-particles formation on the addition of a reducing  
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31 agent. This prompted the arrangement of hydrogel composites with un-collected nano-particles  
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33 (Thoniyot et al., 2015a; Wu et al., 2013). A modified technique of nano-particle synthesis within  
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35 hydrogel matrix in the absence of thiol group was developed by Saravanan et al., (Saravanan et  
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37 al., 2007). Ag nano-particle/polyacrylamide composite was synthesised by free-radical cross-  
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39 linking polymerization using acrylamide and Ag (II) ions aqueous solution. Comparison between  
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41 the non-Ag-loaded polyacrylamide gel and Ag-loaded polyacrylamide nano-composite hydrogels  
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43 showed remarkable enhancement in the properties of swelling along with the electron transfer  
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45 resistance. These properties were found to depend on the amount of Ag nano-particles in the  
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47 hydrogel.  
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### 54 **5.4. Hydrogel nano-composites via Cross-linking**

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4 Hydrogel composites can be synthesized by using semiconductor nano-particle as a cross-linker.  
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6 Zhang et al., prepared semiconductor hydrogel nano-composites by self-polymerization in the  
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8 presence of light (Zhang et al., 2013). In this technique, mainly four parts were used: (a) H<sub>2</sub>O,  
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10 (b) semiconductor nano-particles of zinc oxide, titanium dioxide, Fe<sub>2</sub>O<sub>3</sub>, tin dioxide, cadmium  
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12 selenide or cadmium telluride, all nano-particles were water soluble, (c) N, N-dimethyl  
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14 acrylamide and (d) clay nano-sheets. Semiconductor nano-particles were used to initiate the self-  
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16 polymerization of N, N-dimethyl acrylamide (Paddon et al., 2013). Authors mentioned that  
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18 cadmium selenide and cadmium telluride were capable to synthesize strong hydrogel even in the  
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20 presence of visible light. They also recommended that valuable nano-particles can be  
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22 immobilized in hydrogels for their aggregate application in the sake of joining their mechanical  
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24 and physiochemical properties. Additionally, the declared result for this hydrogel demonstrated  
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26 superb mechanical quality (most noteworthy compressive nature of 4.153 MPa and inflexibility  
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28 1.535 MPa) and high adaptability (most extraordinary protracting of 2784 %).

## 35 **5.5. Hydrogel Composites Formation using Nano-particles, Polymers and Distinct Gelator**

### 36 **Molecules**

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39 Wu et al., 2013 prepared silicon based anodes by adding silicon nano-particles into a conducting  
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41 polymer hydrogel (Wu et al., 2013). A well-developed three-dimensional structure of conducting  
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43 polymer hydrogel with silicon nano-particles was produced through *in situ* polymerization. The  
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45 following interactions were described in the synthesis of this hydrogel: (a) hydrogen bonding  
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47 between silicon surface and phytic acid, (b) electrostatic interaction among silicon surface and  
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49 positively charged aniline polymer. The viscous hydrogel mixture became dark green because of  
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51 phytic acid gelator (C. Wang et al., 2013). Such a hierarchical hydrogel system merges with  
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53 different important characteristics including persistent electrically conductive polyaniline  
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4 network, binding with the Si surface through either the cross-linker hydrogen bonding with  
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6 phytic acid or electrostatic interaction with the positively charged polymer and porous space for  
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8 volume development of Si particles. The developed materials demonstrated cycle life of 5,000  
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10 cycles with more than 90% limits maintenance at current thickness of  $6.0\text{A g}^{-1}$ .  
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## 14 **6. Biodegradability of Hydrogels**

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16 To nullify the environmental effects of solid wastes, bio-degradable polymers are now emerging  
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18 as an alternate for non-biodegradable polymeric materials (Li et al., 2012). For the fabrication of  
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20 any bio-degradable material such as bio-plastics, the material is designed to develop some  
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22 changes in its chemical behaviour under fixed environmental conditions (Kaith et al., 2015). Bio-  
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24 degradability is one of the environment-friendly responses of the hydrogels that ensures its utility  
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26 in target drug delivery applications as hydrogels degraded by simple aqueous breakdown are  
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28 extensively used for drug distribution. However, the hydrogels degraded by enzymes are  
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30 regarded as site specific transporting carriage materials. This is due to the fact that enzymes lie  
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32 over the restricted sites of the body (Bouhadir et al., 2001). Usually, hydrogels are degraded by  
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34 different types of methods (**Figure 3**) for example, a novel approach was developed by Kaith et  
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36 al., using composting method and soil burial to degrade hydrogels within 77 days of contact  
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38 (Kaith et al., 2015). In another work, Jeon et al., developed bio-degradable alginate based photo  
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40 cross-linked hydrogels for tissue engineering application. They have designed the material with  
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42 the ester linkages which was hydrolytically degradable during photo polymerization reaction  
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44 (Jeon et al., 2009). Similarly, bio-degradable oligopeptide cross-linked polymethacrylic acid  
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46 based hydrogel was developed by Knipe et al., which was used to deliver protein in small  
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48 intestine. The enzymatic degradation was catalyzed by the trypsin enzymes present inside the  
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50 small intestine (Knipe et al., 2015). In conclusion, because of better bio-degradability of  
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4 hydrogels, they can be considered as the pioneer material for fulling various targets of the  
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6 medical line.  
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## 8 9 **7. Reusability of Hydrogels**

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11 From the economic point of view, hydrogels are most appropriate as their adsorption limit  
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13 doesn't reduce after a couple of recoveries and reuses (Pakdel and Peighamardoust, 2018). In  
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15 hydrogels, high adsorption limit, fast adsorption rate, high mechanical quality, biodegradability,  
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17 broad pH values and reusability are sensible for significant metals and dye removal. (Pakdel and  
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19 Peighamardoust, 2018). The ordinary reusability of the hydrogel has also been shown  
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21 demonstrated by rehashing the adsorption–desorption procedure (Vivek and Prasad, 2015), while  
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23 keeping up an adequate execution (Chiew et al., 2016). Hydrogel has been found to show an  
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25 incredible degree of extraordinary characteristics for reusability. Upto five adsorption-  
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27 desorption cycle hydrogel were found to give good results (Yi et al., 2018). Reusability of  
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29 adsorbent is one of the fundamental parameter in the functional applications of wastewater  
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31 treatment (Awual et al., 2015, 2014). Yi et al. examined the reusability of sodium  
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33 alginate/polyvinyl alcohol/graphene oxide hydrogel with five cycle's tests (Yi et al., 2018). It  
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35 was concluded from the detailed analysis that the adsorptive property for Cu(II) and UO<sub>2</sub>(II) was  
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37 diminished after the main sorption-desorption cycle and afterward without tremendous  
38  
39 assortment until the fifth cycle, showing that sodium alginate/polyvinyl alcohol/graphene oxide  
40  
41 microspheres can be reused in numerous cycles without basic adversity in its adsorption  
42  
43 characteristics (Yi et al., 2018). Recovery and reusability of the hydrogel were considered  
44  
45 utilizing methyl orange as adsorbate by Balachandran and Edamana in 2015 (Vivek and Prasad,  
46  
47 2015). The approximate time period of 20 minute was reported for desorption process and 97 %  
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49 of the colour was recovered. The execution of the hydrogel for five cycles was incredible (90-98  
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4 %). Chiew et al. have explored the mechanical stability of calcium alginate and alginate-  
5  
6 halloysite nano-composite beads for the removal of Pb(II) ions. Reusability was summarized up  
7  
8 to ten successive cycles and percentage desorption was continued to exist at 94 % for both type  
9  
10 of composite beads (Chiew et al., 2016).  
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### 13 14 **8. Utilization of Hydrogel Products**

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16 Hydrogels have been utilized in the field of target drug delivery applications due to their  
17  
18 distinctive physical properties. The sponge like porous network of hydrogels makes them  
19  
20 compatible for filling multiple types of drugs inside their three dimensional matrix (Vermonden  
21  
22 et al., 2012b). The percentage drug release is managed by the diffusion coefficient of the micro  
23  
24 and macromolecules. Hydrogels have been leading as candidate material for tissue engineering  
25  
26 because of their uncommon structural resemblances with the extracellular matrix (Kim et al.,  
27  
28 2015; Yang et al., 2014). Based on the innovative research worldwide, new opportunities are  
29  
30 coming to conquer the challenges of this field due to their capability to command the surface  
31  
32 morphology and shapes during cell transplantation. It has attended numerous examinations  
33  
34 focused on explorations transporting carriage material for bio-active substances or as a platform  
35  
36 for organizing the cells and to certain the growth of targeted tissues (Kamata et al., 2015;  
37  
38 Seliktar, 2012). In addition to all leading properties of hydrogels, it can also be changed into  
39  
40 defined geometries and purpose of this is to control cell shape and to bring specified biomimetic  
41  
42 design. The physico-chemical properties of the hydrogels can be changed in accordance to the  
43  
44 specified natural cells or tissue by controlling its cross-linking and polymer chemistry in order to  
45  
46 bring out required biological results (Rosales and Anseth, 2016; Uto et al., 2017). The success of  
47  
48 stem cell culture exclusively depends upon targeted differentiation and it is believed that  
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50 homogenous hydrogel surfaces are convenient for the differentiation of stem cells (Higuchi et al.,  
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4 2013). Flexible and soft conductors are valuable parts of bioelectronics that are useful in  
5  
6 biomedical applications. Conductive hydrogels can be effectively used as soft conductors  
7  
8 because of their molecular resemblance to soft tissues and high water absorption ability.  
9  
10 Recently, hydrogels have been utilized in supercapacitors, energy storage, human health  
11  
12 monitoring and electronic skin to name a few (Han et al., 2017; Oh et al., 2016). The adsorption  
13  
14 mechanism of hydrogels is now emerging as feasible option to acquire sustainable approach  
15  
16 towards water treatment mission. The reusability of hydrogels has been revealed by regaining the  
17  
18 adsorption capacity by many consecutive cycles (Khan and Lo, 2016b; Liu et al., 2017) and this  
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20 property makes it as economical material for challenging rest of the developing techniques.  
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## 26 27 **9. Alginate (Natural Polysaccharide)**

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29 Alginate comprises of 30-60% of brown algae. In account of its chemical and physical  
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31 properties, it has been extensively studied due to its ability to form gels and micro-particles (Goh  
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33 et al., 2012; Khotimchenko et al., 2001; Lee and Mooney, 2012; Pawar and Edgar, 2012; Pereira  
34  
35 et al., 2013; Thakur, 2016, 20016). For example, Pereira et al prepared alginate based film for  
36  
37 bio-medical applications. The alginate based hydrogel thin film composed of alginate and aloe  
38  
39 vera gel was used for the treatment of skin. Alginate mixtures are considered to be bio-  
40  
41 compatible, safe, perishable and non-immunogenic (Paques et al., 2014). It can be marked as  
42  
43 anionic biopolymer consisting of mannuronic and guluronic acid units in the irregular blocks  
44  
45 (You et al., 2001). The mannuronic acid forms  $\beta$  (1 $\rightarrow$ 4) linkages and guluronic acid forms  $\alpha$   
46  
47 (1 $\rightarrow$ 4) linkages. The mannuronic and guluronic acid units are joined by glycosidic linkages.  
48  
49 Guluronic acid segments exhibit rigid and bended conformations that provides stiffness to the  
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51 molecular chains (de Vos et al., 2006).  
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### 58 59 **9.1. Alginate Hydrogels**

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4 Alginate hydrogels can be designed via cross-linking the polymer chains. The physical and  
5 chemical properties of the alginate hydrogels are dependent on cross-linking density, type of  
6 cross-linking along with the molecular weight (Franklin and Ohman, 2002). For example, Chan  
7 et al., synthesized alginate cross linked network based on external and internal gelation method.  
8  
9 The prepared sample was utilized for coating and drug delivery of living system (Chan et al.,  
10 2006). One of the suitable method for the formation of alginate hydrogels is inter-molecular  
11 cross-linking in which only guluronic groups of alginate participate with divalent cation such as  
12 calcium (El-Sherbiny, 2010). Numerous divalent cations show distinctive affinity towards  
13 alginate but calcium is more often used for alginate gelation. The properties of alginate  
14 hydrogels such as solubility, hydrophobicity and biological activity can be improved by  
15 modifying alginate through their available hydroxyl and carboxyl groups (Zimmermann et al.,  
16 1992).

## 33 **9.2. Alginate Modification**

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36 Molecular adaptations generally depends on three imperative factors namely reactivity, solubility  
37 and characterization (Pawar and Edgar, 2012a) as shown in **Figure 4**.

### 40 **9.2.1. Solubility**

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43 The dissolvability of alginates in water is represented by three parameters: (a) pH of the  
44 dissolvable, (b) ionic quality of the medium and (c) nearness of gelling particles in the  
45 dissolvable. The solvency of alginates depends emphatically on the condition of the spine  
46 carboxylic acid groups. For derivatization, alginate mixtures can be dissolved in natural aqueous  
47 and organic or combined aqueous-organic media. The possibility of solvent system can impose  
48 the variety of reagents which can be used for modification (Galant et al., 2006).

### 57 **9.2.2. Reactivity**

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4 The two secondary -OH positions (C-2 and C-3) or the one carboxylic group at C-6 position can  
5  
6 be modified in alginate mixtures. The reactivity difference between these two functionalities can  
7  
8 be useful for selective modifications (Donati et al., 2003). Because of little reactivity differences,  
9  
10 selection for the modification is challenging for both positions. The reaction can be controlled in  
11  
12 terms of selective modification of the mannuronic and guluronic block residues. The action of  
13  
14 alginates towards acids, bases and diminishing agents cannot be ignored when performing  
15  
16 derivatization responses. Aggressive degradation responses can cause quick sub-atomic weight  
17  
18 reduction in short periods (Yang et al., 2011).  
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### 23 **9.2.3. Characterization**

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25 In order to realize the chemical substitution configurations in the alginate derivatives, there is  
26  
27 always a need of various alginate samples within a range of mannuronic/guluronic value. For the  
28  
29 detailed understanding of the substitution patterns, (Kim et al., 2000) derivatization of alginate  
30  
31 mixture is required in mannuronic and guluronic blocks. More facilitated analytical techniques  
32  
33 are often required to study the complex nature of the alginate co-polysaccharide backbone.  
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35 Because of the unpredictable nature of the alginate co-polysaccharide backbone, utilization of  
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37 cutting edge investigative methods is most vital.  
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## 43 **10. Sodium Alginate**

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45 Sodium alginate is a product of many unprocessed sugars. It has the capability to be dissolved in  
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47 both cold and hot water with strong stirring. In presence of divalent calcium ion, sodium alginate  
48  
49 builds a gel without any need of heat. In modern food technology, sodium alginate is used to  
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51 create spheres in which sodium alginate is blended with calcium salts (Eghbalifam et al., 2015).  
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53 In food industry, sodium alginate is used to increase viscous nature of liquids, as an emulsifier  
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55 (Mahdavinia et al., 2016) as well as making tasteless indigestion tablets (Park and Kim, 2001).  
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## 11. Sodium Alginate based Hydrogels for Water Purification

Sodium alginate based hydrogels have been used in wastewater treatment (Belhouchat et al., 2017; Bhattacharyya and Ray, 2015; Fan et al., 2013; Fatin-Rouge et al., 2006; Gao et al., 2011; Hosseinzadeh and Abdi, 2017; Karthiga Devi et al., 2016; Lakouraj et al., 2014; Lam et al., 2017; Lu et al., 2015; Mahdavinia et al., 2013; Ngah and Fatinathan, 2008; Rashidzadeh et al., 2015; Ren et al., 2016; Tally and Atassi, 2016; Thakur, 2016; Thakur et al., 2016; Thakur and Arotiba, 2017, n.d.; W. Wang et al., 2013b, 2013a; Wu et al., 2017; Zhuang et al., 2016) (**Table 1**), (**Table 2**). Mahdavinia et al., prepared kappa-carrageenan-sodium alginate/montmorillonite (CarAlg/MMt) hydrogel nano-composite by using acrylamide as monomer (Mahdavinia et al., 2013). The synthesized hydrogel nano-composite was utilized for the adsorption of cationic crystal violet dye from water. The maximum adsorption capacity obtained was found to be 88.8 mg g<sup>-1</sup>. The basic Na-montmorillonite (Na-MMt) clay and hydrogel nano-composite containing 0.25 g and 0.5 g of basic Na-MMt (CarAlg/MMt0.25, CarAlg/MMt0.5) were characterized by XRD. A sharp diffraction peak at  $2\theta = 7.6^\circ$  with d-spacing of 11.61 Å confirmed the basic Na-MMt. The peak at  $2\theta = 7.6^\circ$  vanished due to the intercalation of Na-MMt clay in the matrix of nano-composite. In the TEM image of CarAlg/MMt0.25, the black and fine lines correspond to Na-MMt layers. The surface of hydrogel without clay (CarAlg) was found to be smooth and tight. The surface became porous after the addition of Na-MMt nanoclay into the hydrogel. The adsorption kinetics was used to measure the expected equilibrium time for adsorption of crystal violet dye (Mahdavinia et al., 2013). The adsorption of crystal violet dye was sharply increased at the beginning and then became constant. The equilibrium time of 2 hours was reported for dye adsorption. The dye adsorption capacity was increased by adding Na-MMt clay in hydrogel. The rate of adsorption of dye was increased with the amount of clay in hydrogel. The formation of

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4 active centers because of porosity was the cause of increase in speed of crystal violet adsorption  
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6 by the hydrogel nano-composite (Wang et al., 2008). The maximum adsorption capacity of 88.8  
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8 mg g<sup>-1</sup> was achieved by the CarAlg/MMt0.5 sample.  
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10  
11 Sodium alginate-g-poly(acrylic acid-co-acryl amide)/clinoptilolite hydrogel nano-composite was  
12  
13 synthesized through free radical polymerization technique (Rashidzadeh et al., 2015). The  
14  
15 developed hydrogel nano-composite was utilized for the removal of methylene blue dye and  
16  
17 94.34 mg g<sup>-1</sup> was the maximum adsorption capacity. FTIR spectrum of sodium alginate-g-  
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19 poly(acrylic acid-co-acryl amide)/clinoptilolite, sodium alginate-g-poly(acrylic acid-co-acryl  
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21 amide) and methylene blue adsorbed sodium alginate-g-poly(acrylic acid-co-acryl  
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23 amide) and methylene blue adsorbed sodium alginate-g-poly(acrylic acid-co-acryl  
24  
25 amide)/clinoptilolite are given in **Figure 5a-c**. In **Figure 5a** and **5b**, the absorption bands at 1650  
26  
27 cm<sup>-1</sup> and 1450 cm<sup>-1</sup> were due to sodium alginate and carboxylate stretching respectively. Broad  
28  
29 band at 3000-3500 cm<sup>-1</sup> was due to the stretching of hydroxyl groups of the polysaccharide. The  
30  
31 shift in the -COOH, -OH adsorption band in the spectrum of methylene blue adsorbed sodium  
32  
33 alginate-g-poly(acrylic acid-co-acryl amide)/clinoptilolite (**Figure 5c**) indicated the adsorption of  
34  
35 methylene blue dye onto sodium alginate-g-poly(acrylic acid-co-acryl amide)/clinoptilolite  
36  
37 hydrogel nano-composite. The sodium alginate-g-poly(acrylic acid-co-acryl amide)/clinoptilolite  
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39 hydrogel nano-composite was composed of pores with size of ten microns as indicated in SEM  
40  
41 morphology (**Figure 5d**). Adsorption mechanism was explained on the basis of change in colour  
42  
43 of sodium alginate-g-poly(acrylic acid-co-acryl amide) hydrogel (**Figure 6a,c**) and sodium  
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45 alginate-g-poly(acrylic acid-co-acryl amide)/clinoptilolite hydrogel nano-composite (**Figure**  
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47 **6b,d**) after the adsorption of methylene blue dye. The electrostatic attraction between amide and  
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49 carboxylic of sodium alginate-g-poly(acrylic acid-co-acryl amide)/clinoptilolite hydrogel nano-  
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51 composite and imine groups of methylene blue dye was responsible for adsorption of dye.  
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4 Wu et al., prepared sodium alginate/ $\beta$ -cyclodextrin/graphene oxide hydrogel nano-composite  
5 adsorbent for the adsorption of methylene blue dye (Wu et al., 2017). The graphene oxide used  
6 for the synthesis of hydrogel nano-composite was produced by an improved Hummers method.  
7 The highest adsorption capacity of synthesized adsorbent was 133.24 mg g<sup>-1</sup>. The thickness of 1  
8 to 2 nm and micro meter-sized planes of graphene oxide was confirmed by atomic force  
9 microscopy analysis (**Figure 7a**). TEM image of graphene oxide with broken surface is shown in  
10 **Figure 7f**. The colour of sodium alginate/ $\beta$ -cyclodextrin/graphene oxide hydrogel nano-  
11 composite was faint yellow whereas colour of methylene blue loaded sodium alginate/ $\beta$ -  
12 cyclodextrin/graphene oxide hydrogel nano-composite was dark blue black (**Figure 7b**). The  
13 faint yellow colour of sodium alginate/ $\beta$ -cyclodextrin/graphene oxide hydrogel nano-composite  
14 was due to the presence of graphene oxide. The porous and honeycomb-like structure of sodium  
15 alginate/ $\beta$ -cyclodextrin/graphene oxide adsorbent was confirmed by SEM morphology (**Figure**  
16 **7c-e**). The adsorption of methylene blue onto sodium alginate/ $\beta$ -cyclodextrin/graphene oxide  
17 hydrogel nano-composite was explained via hydrogen bonding,  $\pi$ - $\pi$  interaction and electrostatic  
18 attraction (Heidarizad and Şengör, 2016) as represented in **Figure 8**.

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41 Thakur et al., prepared titania incorporated sodium alginate/acrylic acid hydrogel nano-  
42 composite for the adsorption of methylene blue dye from aqueous solution (Thakur et al., 2016).  
43 The reported maximum adsorption capacity was 2257.36 mg g<sup>-1</sup>. The synthesis of hydrogel  
44 nano-composite was explained by free radical polymerization of acrylic acid onto sodium  
45 alginate using potassium persulphate and N,N'-methylenebisacrylamide as initiator and cross-  
46 linker respectively. The adsorption mechanism of methylene blue dye was explained by  
47 electrostatic attraction (between TiO<sub>2</sub> or COO<sup>-</sup> and cationic dye) and hydrogen bonding  
48 (between OH and imines groups of dye) (Pourjavadi et al., 2014) (**Figure 9**).  
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4 Ternary hydrogel nano-composite of graphene oxide/sodium alginate/polyacrylamide has been  
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6 synthesized for the adsorption of cationic as well as anionic dyes (Fan et al., 2013). The  
7  
8 developed ternary hydrogel nano-composite was ionically cross-linked by using Ca(II) ion. On  
9  
10 applying stress, the pure polyacrylamide hydrogel was destroyed (**Figure 10a-c**). However,  
11  
12 graphene oxide/sodium alginate/polyacrylamide hydrogel nano-composite was not destroyed on  
13  
14 applying stress (**Figure 10d-f**). On the removal of stress, graphene  
15  
16 oxide/sodiumalginate/polyacrylamide hydrogel nano-composite regained its original shape. The  
17  
18 possible scheme for graphene oxide/sodium alginate/polyacrylamide ternary hydrogel nano-  
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20 composite is shown in **Figure 10g**. Intercalation of layered structure graphene oxide with  
21  
22 hydrogel network was presented. The mechanical stability of hydrogel nano-composite was  
23  
24 improved by the exchange of load among graphene oxide nanosheets and polymer chains.  
25  
26 Ionically cross-linked as well as covalently cross-linked polymers were presented in hydrogel  
27  
28 network. Also, polymer chains of hydrogel interacted with graphene oxide nanosheets through  
29  
30 hydrogen bonding. The sponge-like morphology was reported for hydrogel samples as shown in  
31  
32 **Figure 11**. The structure of polyacrylamide and sodium alginate/polyacrylamide hydrogels  
33  
34 contained holes with uniform size (**Figure 11a-c**). The lamellar and dense structures were  
35  
36 formed after the addition of graphene oxide into the hydrogel network (**Figure 11d-f**). The  
37  
38 change in colour of different dyes after the adsorption by sodium alginate/polyacrylamide  
39  
40 hydrogel nano-composite is represented in **Figure 12**. The following interactions were used to  
41  
42 explain the adsorption mechanism: (a) amino and azo functional group of dye with carboxylic  
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44 functional groups of hydrogel nano-composite, (b)  $\pi$ - $\pi$  interactions between aromatic groups.  
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56 Sodium alginate based silver hydrogel nano-composite was developed by one step green method  
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58 (Karthiga Devi et al., 2016). Devi et al., have used this hydrogel nano-composite for methylene  
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4 blue dye removal and  $213.7 \text{ mg g}^{-1}$  was the maximum adsorption capacity reported. Synthesis of  
5  
6 silver nano-particles was performed by reduction with *M. maderaspatna* extract. Silver nano-  
7  
8 particles were characterized by using UV-vis-spectrophotometric analysis. The band at 443 nm  
9  
10 confirmed the formation of silver nano-particles (A Bhosale and M Bhanage, 2015). Hydrogel  
11  
12 beads were synthesized by gelation process of sodium alginate and carbopol 937. Finally, silver  
13  
14 nano-particles introduced into the hydrogel beads led to the formation of silver nano-composite  
15  
16 for dye adsorption studies. The crystalline structure and size of silver nano-composite was  
17  
18 analyzed by XRD, diffraction peaks at  $28^\circ$ ,  $32^\circ$ ,  $38^\circ$ ,  $46^\circ$ ,  $54^\circ$ ,  $57^\circ$ ,  $76^\circ$  with planes (1 1 1), (2 0  
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20 0), (2 1 0), (2 2 0), (3 1 1), (2 2 2), (4 2 0) corresponded to crystalline silver nano-particles. TEM  
21  
22 analysis was used to verify the size and shape of the silver nano-particles containing carbopol-  
23  
24 alginate hydrogel beads. The reported average crystal size was about 19.3 nm with uniform  
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26 shape.  
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34 The emulsion micelle has been incorporated into the vinyl-functionalized sodium alginate-co-  
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36 methyl acrylic acid-co-italic acid hydrogel for the formation of nano-porous hydrogel (W. Wang  
37  
38 et al., 2013b). This synthesized nano-porous hydrogel was used for the removal of Pb(II) ion  
39  
40 from aqueous solution. Three dimensional network of vinyl-functionalized sodium alginate-co-  
41  
42 methyl acrylic acid-co-italic acid hydrogel was developed through free radical polymerization  
43  
44 reaction (**Figure 13**). The reaction between sodium alginate and glycidyl methacrylate led to the  
45  
46 introduction of vinyl groups on sodium alginate. The sulfate anion radicals were produced by  
47  
48 hemolytic breakage of ammonium persulfate (Buchholz and Graham, 1998). The chain  
49  
50 propagation step was carried out in between radicals, vinyl-functionalized sodium alginate chain  
51  
52 and monomers. Different chains of sodium alginate were cross-linked through methyl acrylic  
53  
54 acid and italic acid polymer chains. Emulsion micelles were formed by sodium dodecyl sulfonate  
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4 and n-Octane, which were introduced into the cross-linked hydrogel network. Nano porous  
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6 hydrogel was formed after the removal of emulsion micelles as shown in **Figure 13**. The peaks  
7  
8 at  $\delta$  3.55–4.01 and  $\delta$  4.86 were due to the non-anomeric and anomeric protons in the  $^1\text{H}$  NMR  
9  
10 spectrum of sodium alginate (**Figure 14a**). New peaks were represented in the vinyl-  
11  
12 functionalized sodium alginates hydrogel due to presence of methyl protons and methacryloyl  
13  
14 double bonded hydrogen's at  $\delta$ 1.80 ppm and  $\delta$  5.60,  $\delta$ 6.02 ppm, respectively. In  $^{13}\text{C}$  NMR  
15  
16 spectrum of sodium alginate, peaks in the range of  $\delta$ 63.5–102.18 ppm were due to the sugar  
17  
18 carbons of sodium alginate (**Figure 14b**). The following new peaks were observed in the  $^{13}\text{C}$   
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20 NMR spectrum of vinyl-functionalized sodium alginate: (a)  $\delta$  19.46 ppm due to methyl carbon,  
21  
22 (b)  $\delta$ 129.28 and  $\delta$ 138.31 ppm assigned to vinyl carbons, (c)  $\delta$ 171.86 ppm due to carbonyl  
23  
24 carbon. Hence, modification of sodium alginate with glycidyl methacrylate and incorporation of  
25  
26 vinyl groups on the sodium alginate matrix were confirmed by these NMR results. The  $^1\text{H}$  NMR  
27  
28 and  $^{13}\text{C}$  NMR spectrum of vinyl-functionalized sodium alginate was identical to modified cashew  
29  
30 gum with glycidyl methacrylate, indicating mechanism of modification for sodium alginate and  
31  
32 cashew gum with glycidyl methacrylate was identical (Guilherme et al., 2005). Ring opening  
33  
34 mechanism was believed to take place between sodium alginate with glycidyl methacrylate<sup>91</sup>.  
35  
36 The smooth and dense surface without any pore was observed in the morphology of vinyl-  
37  
38 functionalized sodium alginate-co-methyl acrylic acid-co-italic acid hydrogel (**Figure 15a**).  
39  
40 However, structure with uniform pores (20–60 nm) was obtained after the addition of emulsion  
41  
42 micelle into the vinyl-functionalized sodium alginate-co-methyl acrylic acid-co-italic acid  
43  
44 (**Figure 15b-d**).

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46 Sodium alginate-g-polyacrylic acid/polyvinylpyrrolidone/gelatin granular hydrogel was  
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48 synthesized through grafting and cross-linking using gelatin, acrylic acid and  
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4 polyvinylpyrrolidone (W. Wang et al., 2013a). The developed sodium alginate-g-polyacrylic  
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6 acid/polyvinylpyrrolidone/gelatin granular hydrogel was utilized for the adsorption of heavy  
7  
8 metal ions. The adsorption capacity for Ni(II), Cu(II), Zn(II) and Cd(II) was found to be 3.028  
9  
10 mmol g<sup>-1</sup>, 3.146 mmol g<sup>-1</sup>, 2.911 mmol g<sup>-1</sup> and 2.862 mmol g<sup>-1</sup> respectively. The sodium  
11  
12 alginate-g-polyacrylic acid/polyvinylpyrrolidone/gelatin granular hydrogel was prepared by  
13  
14 chemical and physical reactions (Pourjavadi et al., 2008) (**Figure 16**). The H<sup>+</sup> of acrylic acid was  
15  
16 used to generate electro positivity on polyvinylpyrrolidone and gelatin. Hence,  
17  
18 polyvinylpyrrolidone and gelatin were interacted with sodium alginate and grafted polyacrylic  
19  
20 chains through electrostatic and hydrogen-bonding. These interactions among  
21  
22 polyvinylpyrrolidone, gelatin, sodium alginate and acrylic acid led to formation of granular  
23  
24 hydrogel. **Figure 17a** shows the gel like photograph of sodium alginate-g-polyacrylic acid  
25  
26 hydrogel. After the addition of polyvinylpyrrolidone and gelatin, shape of sodium alginate-g-  
27  
28 polyacrylic acid hydrogel was converted into the granular (**Figure 17b**). This granular shape of  
29  
30 hydrogel was maintained in the dry state (**Figure 17c**). Sodium alginate-g-polyacrylic acid  
31  
32 hydrogel exhibited smooth surface (**Figure 17d**), whereas sodium alginate-g-polyacrylic  
33  
34 acid/polyvinylpyrrolidone/gelatin granular hydrogel displayed rough and porous surface (**Figure**  
35  
36 **17e**). Adsorption capacity and recovery capability of the sodium alginate-g-polyacrylic  
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38 acid/polyvinylpyrrolidone/gelatin granular hydrogel for various metal ions were investigated.  
39  
40 The adsorption capacity of sodium alginate-g-polyacrylic acid/polyvinylpyrrolidone/gelatin  
41  
42 granular hydrogel for Cu(II) ion was the highest. The recovery ability of sodium alginate-g-  
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44 polyacrylic acid/polyvinylpyrrolidone/gelatin granular hydrogel was increased with increased in  
45  
46 the hydrogel dosage. For 2.8 g L<sup>-1</sup> dosage, reported recovery percentages were 90.92 (Cu(II)),  
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48 87.70 (Ni(II)), 87.47 (Zn(II)) and 85.55 (Cd(II)).  
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4 Gao et al., have produced hydrogel composite composed of sodium alginate, sodium acrylate and  
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6 medical stone (Gao et al., 2011). Various heavy metal ions (Ni(II), Cu(II), Zn(II) and Cd(II))  
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8 were removed by using sodium alginate-g-sodium acrylate/medical stone hydrogel composite. A  
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10 smooth and non-porous surface was shown by sodium alginate-g-sodium acrylate hydrogel  
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12 (**Figure 18a**) whereas medical stone exhibited rough and sheet surface (**Figure 18b**). The rough  
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14 surface with pores and gap was formed on the addition of medical stone into sodium alginate-g-  
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16 sodium acrylate hydrogen (**Figure 18c**). It was analyzed that surface roughness was increased  
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18 with increase in medical stone amount in sodium alginate-g-sodium acrylate/medical stone  
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20 hydrogel composite (**Figure 18d**). Equilibrium heavy metal ions adsorption capacities for active  
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22 carbon, medical stone and hydrogel composite with different amounts of medical stone were  
23  
24 examined. The adsorption capacities for hydrogel composite with different amount of medical  
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26 stone were found to be higher in comparison to active carbon and medical stone. Many  
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28 carboxylate groups in hydrogel composite were found to form complexes with heavy metal ions  
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30 during adsorption process.  
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39 Alginate/reduced graphene oxide double network hydrogel nano-composite was developed using  
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41 facile method by Zhuang et al. (Zhuang et al., 2016). An adsorption capacity of  $169.5 \text{ mg g}^{-1}$  and  
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43  $72.5 \text{ mg g}^{-1}$  was reported for Cu(II) and  $\text{Cr}_2\text{O}_7^{2-}$  respectively by utilizing alginate/reduced  
44  
45 graphene oxide double network hydrogel nano-composite. The schematic representation of  
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47 hydrogel synthesis is presented in **Figure 19**. The reduced graphene oxide was formed by  
48  
49 reduction of graphene oxide and alginate beads were developed through the addition of sodium  
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51 alginate liquid into the  $\text{CaCl}_2$  solution. Three dimensional networks of alginate and two  
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53 dimensional networks of graphene oxide were produced from the alginate/graphene composite.  
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58 Another three dimensional interpenetrates networks were formed by reduced graphene oxide,  
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4 which led to the formation of double network (**Figure 20**). Many particles like morphologies  
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6 were observed in the SEM image of graphene–alginate single networks (**Figure 20a**), while  
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8 alginate/reduced graphene oxide double-network has shown petal-like morphology (**Figure 20b**).  
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10 The particle morphology of graphene–alginate single networks was not in accordance with  
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12 oxygen element distribution (**Figure 20c**) whereas morphology of alginate/reduced graphene  
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14 oxide double-network was in accordance with oxygen distribution (**Figure 20d**). Hence, it was  
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16 concluded that hydrogen bonds played a crucial role in the production of three dimensional  
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18 networks by reduced graphene oxide. The hydrogen bonding between reduced graphene oxide  
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20 and alginate led to formation of uniform structure of graphene–alginate single networks, while  
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22 hydrogen bonding among the sheets of graphene led to the development of three dimensional  
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24 networks in reduced graphene oxide. In this work, the researchers have concluded that double  
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26 network structure in prepared hydrogel composite enhanced the adsorption capacity as well as  
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28 provided the evidences of repeated usage after many cycles of chemisorptions.  
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36 Tally et al., have synthesized sodium alginate-g-poly(acrylic acid-co-acrylamide) hydrogel by  
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38 utilizing microwave oven (Tally and Atassi, 2016). An adsorption capacity of  $480.77 \text{ mg g}^{-1}$  has  
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40 been reported for Pb(II) ion removal using sodium alginate-g-poly(acrylic acid-co-  
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42 acrylamide)hydrogel. Adsorption of Cd(II), Ni(II) and Cu(II) by sodium alginate-g-poly(acrylic  
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44 acid-co-acrylamide) hydrogel has been also analyzed. Sodium n-dodecyl benzenesulfonate was  
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46 used to produce pore forming micelles in sodium alginate-g-poly(acrylic acid-co-acrylamide)  
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48 hydrogel. Spherical micelles were formed by sodium n-dodecyl benzenesulfonate at fixed  
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50 concentration, which were introduced in the sodium alginate-g-poly(acrylic acid-co-acrylamide)  
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52 hydrogel network. After washing, sodium n-dodecyl benzenesulfonate micelles were removed,  
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54 which led to the formation of pores in sodium alginate-g-poly(acrylic acid-co-acrylamide)  
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4 hydrogel (**Figure 21**). **Figure 22** represents XRD spectra for alginate, poly(acrylic acid-co-  
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6 acrylamide) and sodium alginate-g-poly(acrylic acid-co-acrylamide) hydrogel. The crystalline  
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8 nature was decreased on grafting of copolymer on sodium alginate. This behavior was affirmed  
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10 from the broader peak at  $17^{\circ}$ – $37^{\circ}$  in the XRD spectra of sodium alginate-g-poly(acrylic acid-co-  
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12 acrylamide) hydrogel (**Figure 22**).  
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16 In another study, blending was used to develop sodium alginate-carboxymethyl cellulose gel  
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18 beads (Ren et al., 2016). The synthesized gel bead was used to remove Pb(II) ion from aqueous  
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20 solution. Adsorption percentage of more than 99 was reported for Pb(II) ion by using sodium  
21  
22 alginate-carboxymethyl cellulose gel bead. The wrinkled and low porosity structure was  
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24 exhibited by the pure sodium alginate gel bead (**Figure 23a**), whereas rough and highly porous  
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26 structure was shown by sodium alginate-carboxymethyl cellulose gel bead (**Figure 23b**). High  
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28 surface area and porosity was displayed by cross section of sodium alginate-carboxymethyl  
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30 cellulose gel bead (**Figure 23c**). Internal network of sodium alginate-carboxymethyl cellulose  
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32 gel bead developed through the condensation reaction among hydroxyls groups of  
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34 carboxymethyl cellulose and carboxyl groups of sodium alginate. Sodium alginate-  
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36 carboxymethyl cellulose gel bead was found to be more efficient than cation exchange resin and  
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38 activated carbon for lead ions removal. The lead ion removal percentages were 99.6 %, 74.8 %  
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40 and 66.4 % for sodium alginate-carboxymethyl cellulose gel bead, cation exchange resin and  
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42 activated carbon respectively.  
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## 50 **12. Conclusion**

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54 In this article, we have reviewed a versatile class of sodium alginate based hydrogels that are  
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56 potentially useful for creating multitasking systems intended for achieving advanced adsorption  
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58 properties. Hydrogels integrated with nano-particles, which not only increase the mechanical  
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4 characteristics of the hydrogels but also modulate its adsorption behavior, appear more  
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6 efficiently. Hydrogel nano-composite materials manifest stimuli-responsive properties including  
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8 photo thermal activity, catalysis, target drug delivery, anti-microbial property, drug degradation,  
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10 removal of aquatic pollutants, bio-sensing and many more which makes it ideal for “smart”  
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12 material. Our prime emphasis has been to bring potential design parameters useful for adsorption  
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14 of pollutants. The systematic investigation on hydrogel composites for water purification  
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16 application can persuade more research efforts to label the challenges and upgrade an area of  
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18 interest for the general study of hydrogels. There is still a need for further investigations  
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20 targeting on the intrinsic interactions of nano-particles with polymer matrix in order to alter their  
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22 properties to explore more scientific research.  
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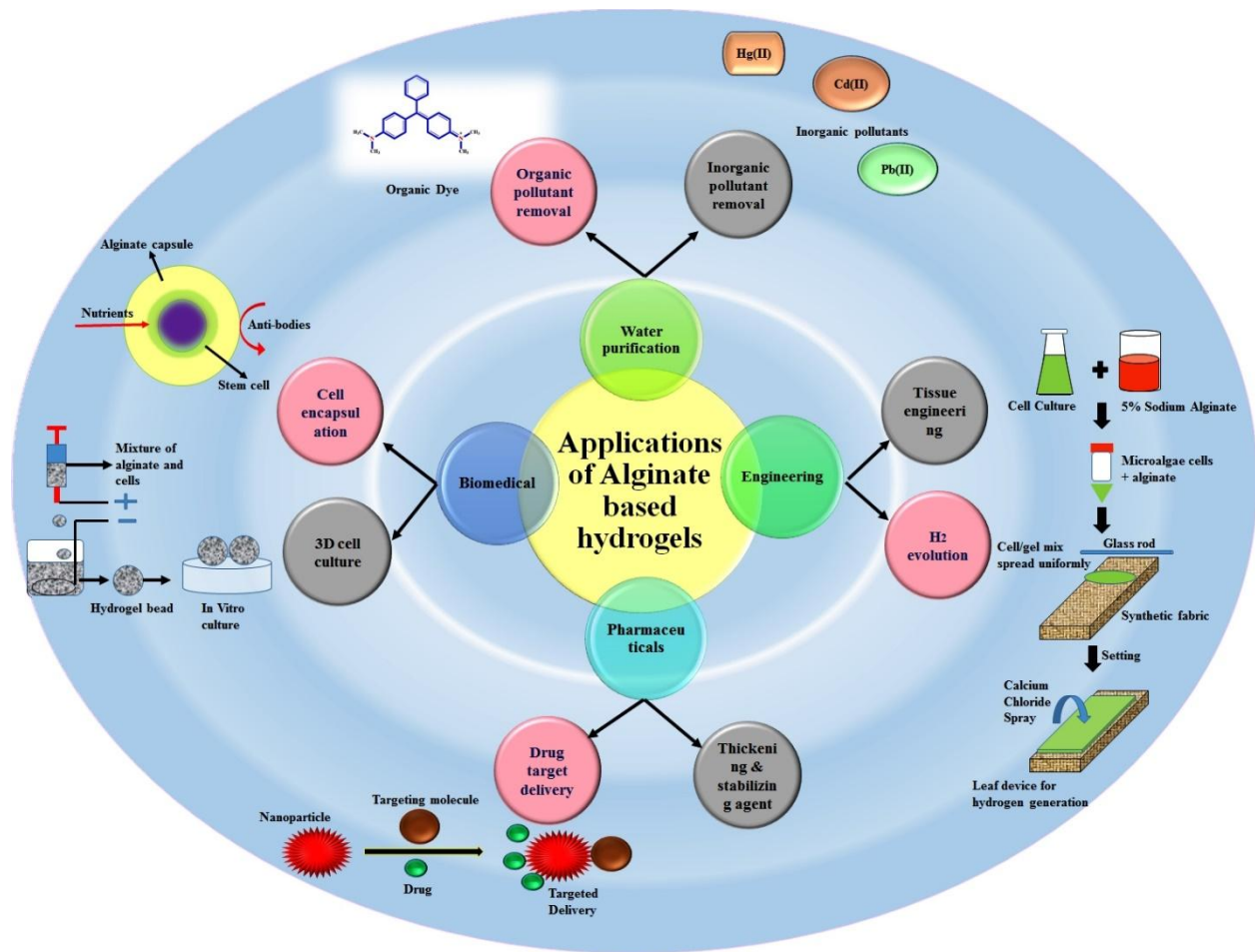
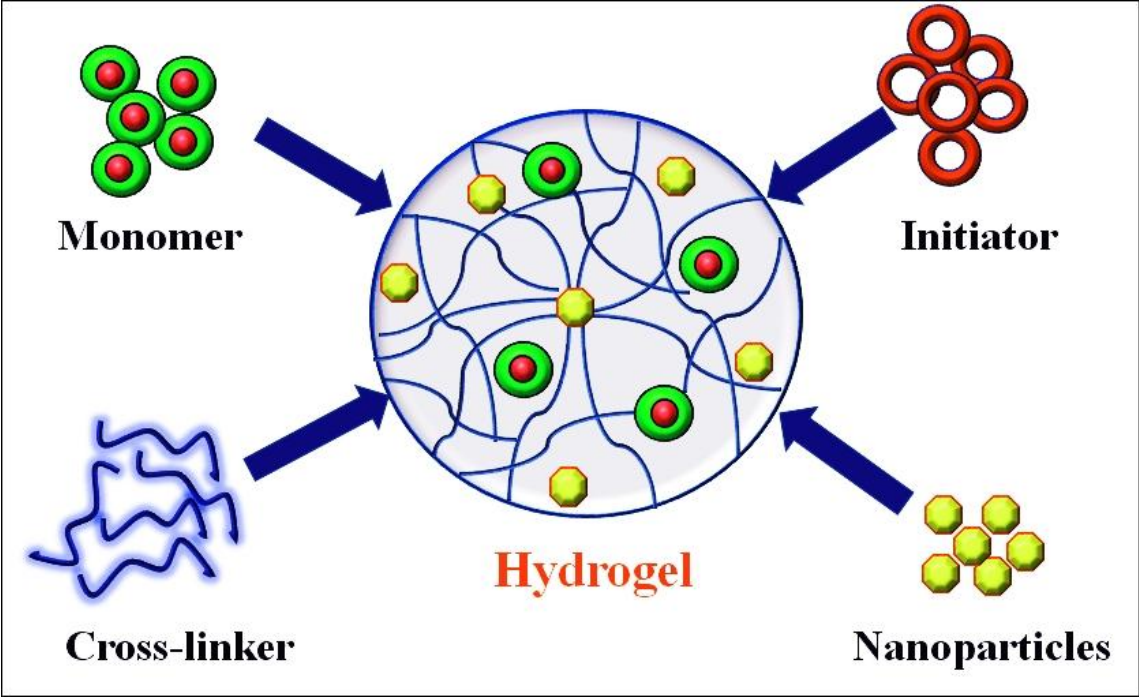


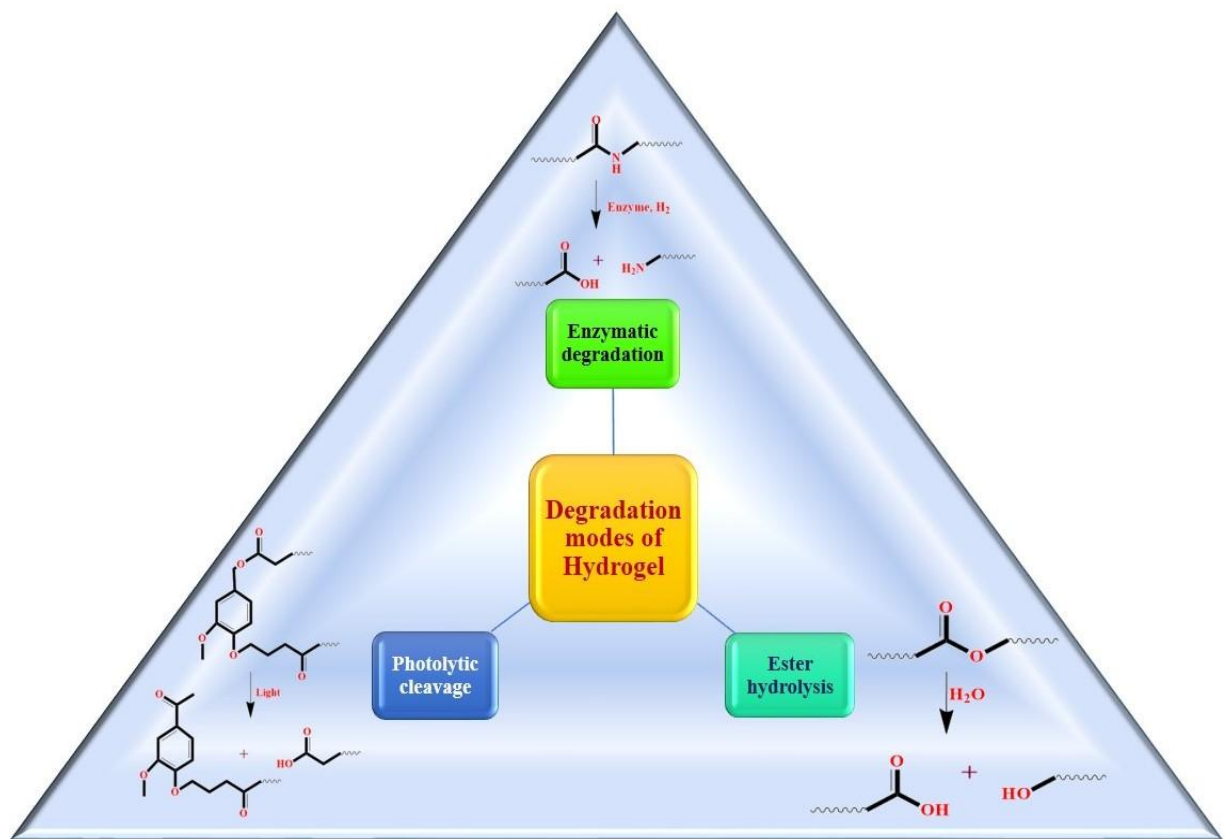
Figure 1. Various applied applications of alginate based hydrogels

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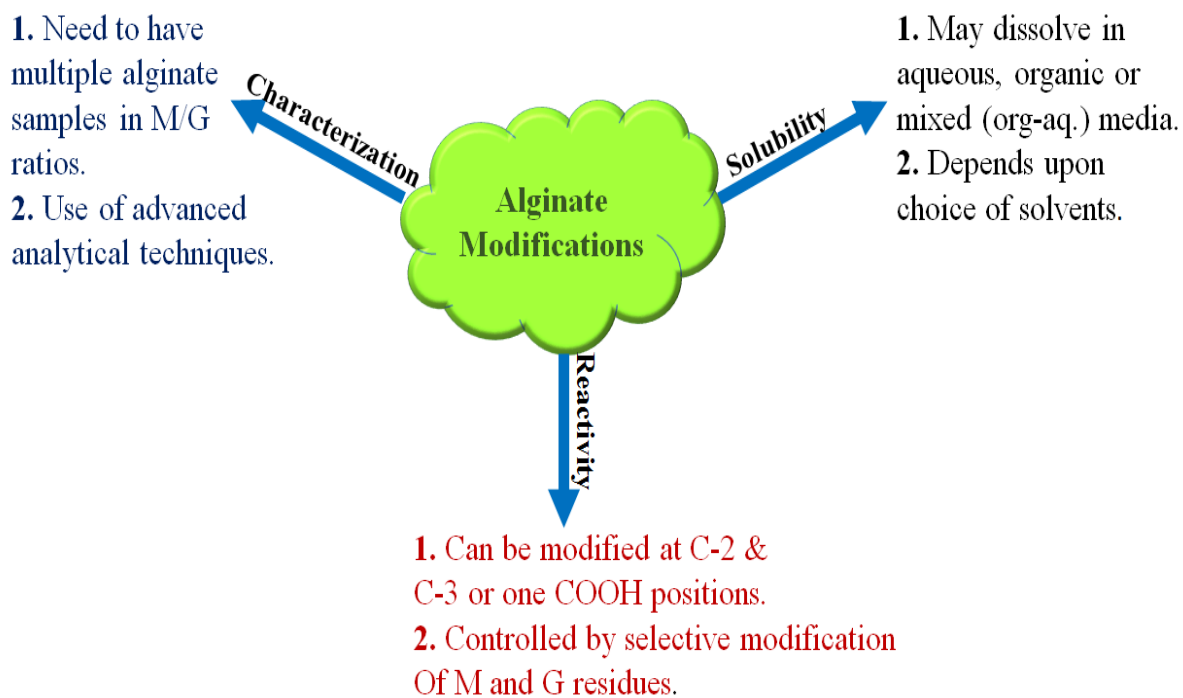
**Figure 2. Schematic diagram of hydrogel preparation.**

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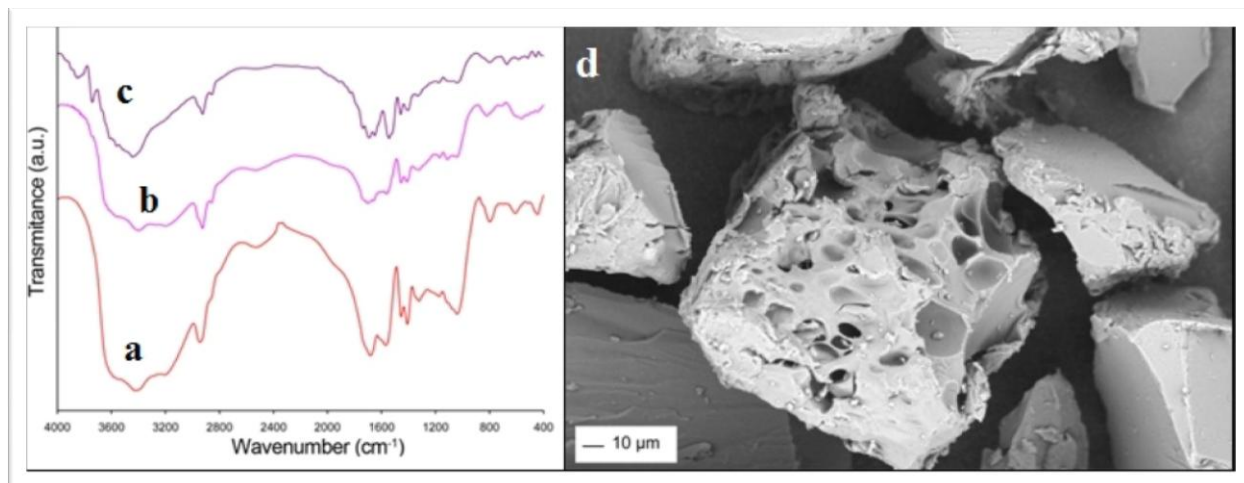


**Figure 3. Different modes of degradation of hydrogel.**

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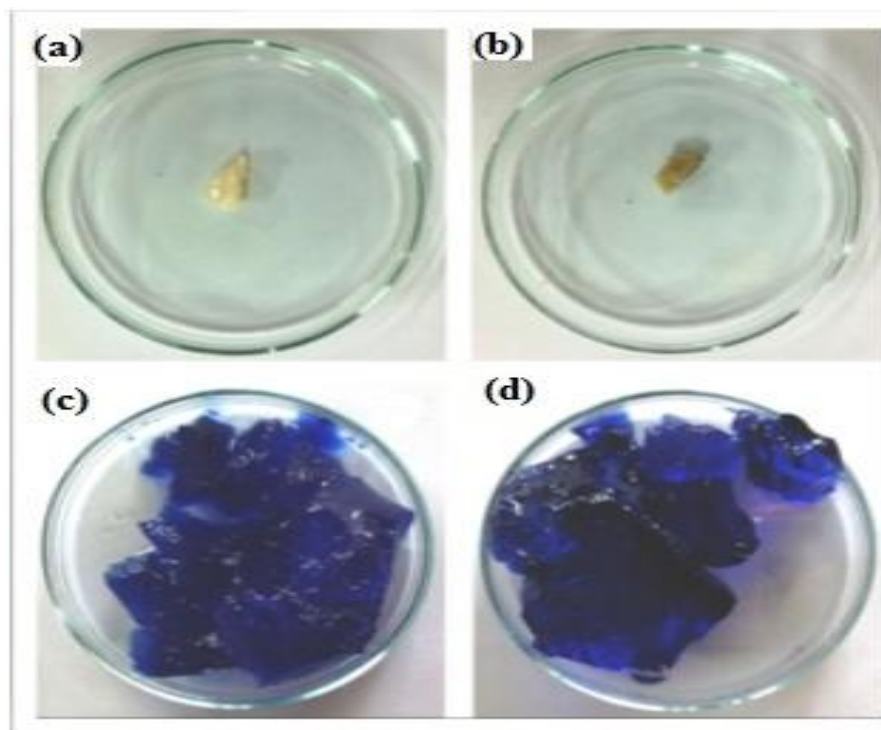


**Figure 4. Parameters governing alginate derivatization. Adopted from (Pawar and Edgar, 2012).**



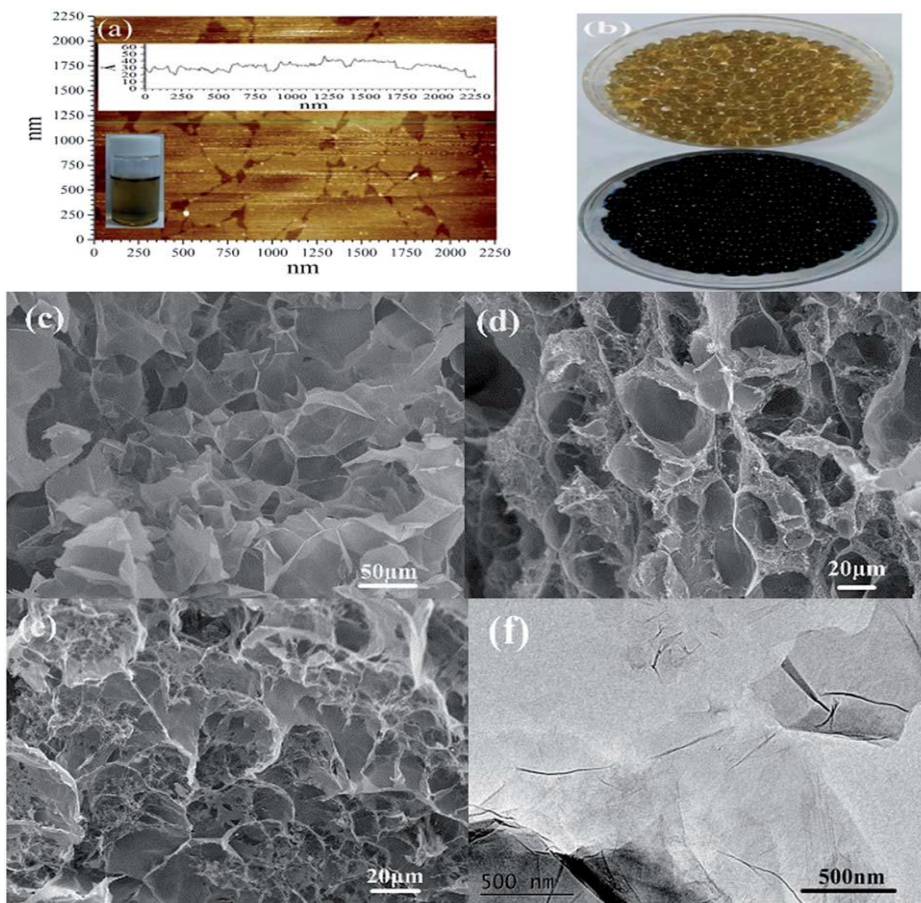
**Figure 5. FTIR spectrum (a) sodium alginate-g-poly(acrylic acid-co-acrylamide)/clinoptilolite, (b) sodium alginate-g-poly(acrylic acid-co-acrylamide) and (c) methylene blue adsorbed sodium alginate-g-poly(acrylic acid-co-acrylamide)/clinoptilolite, (d) SEM morphology of sodium alginate-g-poly(acrylic acid-co-acrylamide)/clinoptilolite (Rashidzadeh et al., 2015). Reprinted with permission from (Rashidzadeh et al., 2015). Copyright 2015 Springer.**



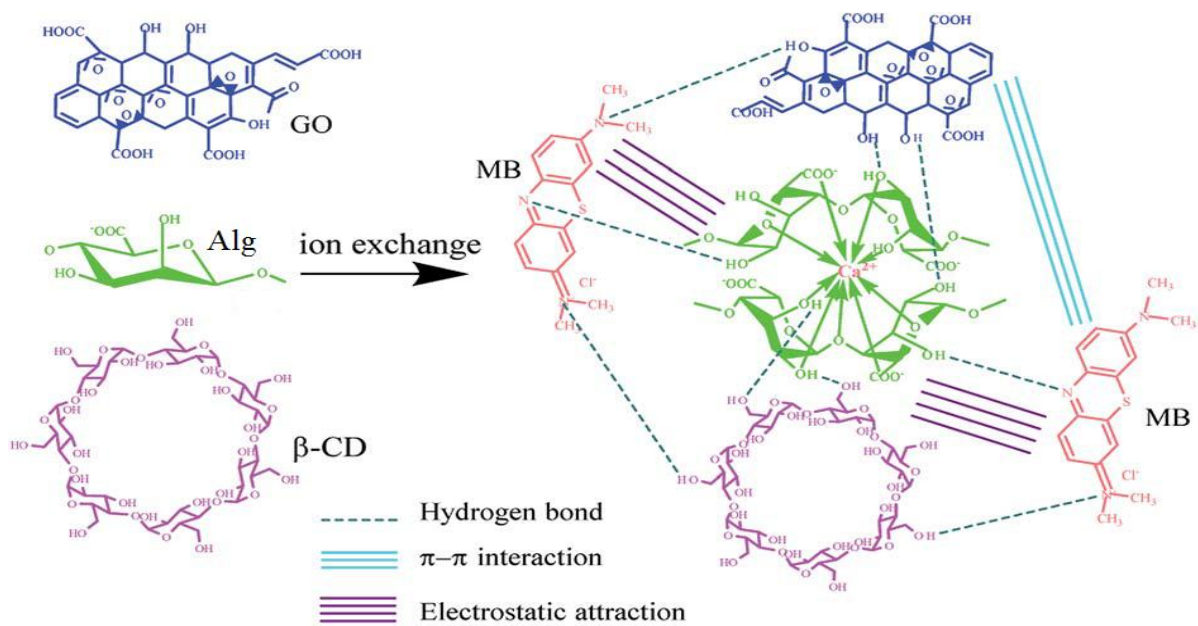


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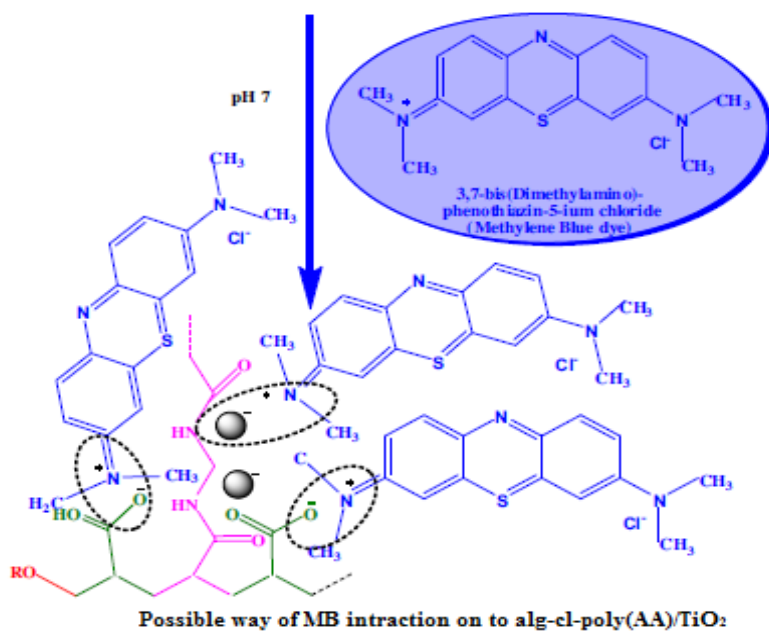
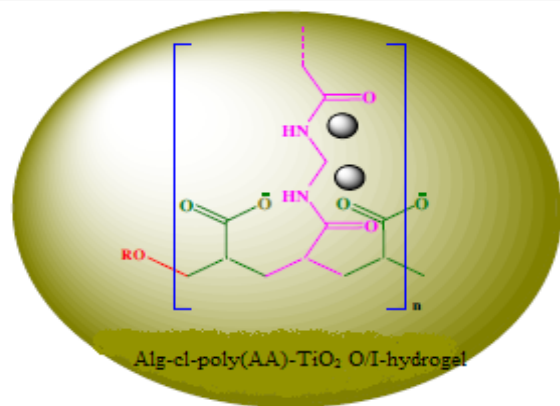
**Figure 6. Images of sodium alginate-g-poly(acrylic acid-co-acryl amide) hydrogel and sodium alginate-g-poly(acrylic acid-co-acryl amide)/clinoptilolite hydrogel nano-composite samples before (a, b) adsorption of methylene blue dye and after (c, d) adsorption of methylene blue dye solution (Rashidzadeh et al., 2015). Reprinted with permission from (Rashidzadeh et al., 2015). Copyright 2015 Springer.**



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37 **Figure 7. (a) Atomic force microscopy image of graphene oxide, (b) image of sodium**  
38 **alginate/ $\beta$ -cyclodextrin/graphene oxide hydrogel nano-composite and methylene blue**  
39 **loaded sodium alginate/ $\beta$ -cyclodextrin/graphene oxide hydrogel nano-composite, (c-e) SEM**  
40 **images of sodium alginate/ $\beta$ -cyclodextrin/graphene oxide hydrogel nano-composite and (f)**  
41 **TEM image of graphene oxide (Wu et al., 2017). Reproduced from (Wu et al., 2017).**  
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**Figure 8. Proposed mechanism for methylene blue adsorption onto sodium alginate/β-cyclodextrin/graphene oxide hydrogel nano-composite (Wu et al., 2017). Reproduced from (Wu et al., 2017). Published by Royal Society of Chemistry.**



**Figure 9. Possible mechanism for interaction of cationic methylene blue (MB) dye with titania incorporated sodium alginate cross-linked acrylic acid hydrogel nano-composite (Alg-cl-poly(AA)-TiO<sub>2</sub>) (Thakur et al., 2016). Reprinted with permission from (Thakur et al., 2016). Copyright 2016 Elsevier.**

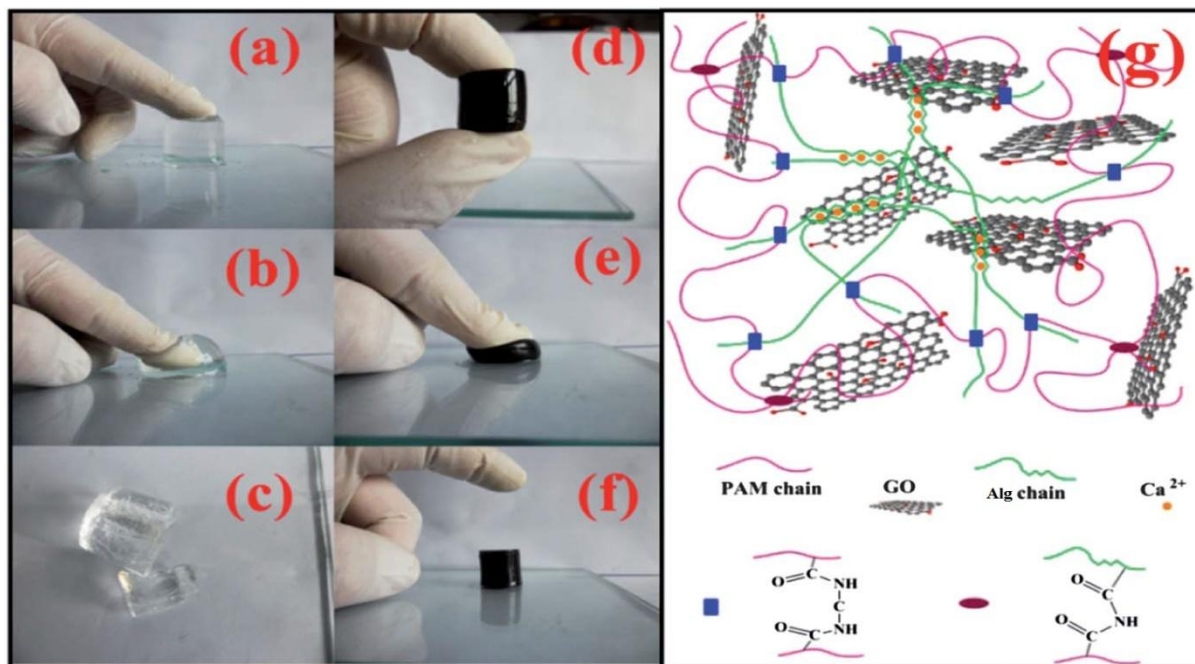


Figure 10. (a-c) Photographs of pure polyacrylamide hydrogel under compression, (d-f) photographs of graphene oxide/sodium alginate/polyacrylamide hydrogel nano-composite under compression, (g) general representation for graphene oxide/sodium alginate/polyacrylamide ternary hydrogel nano-composite (Fan et al., 2013). Reprinted with permission from (Fan et al., 2013). Copyright 2013 Royal Society of Chemistry.



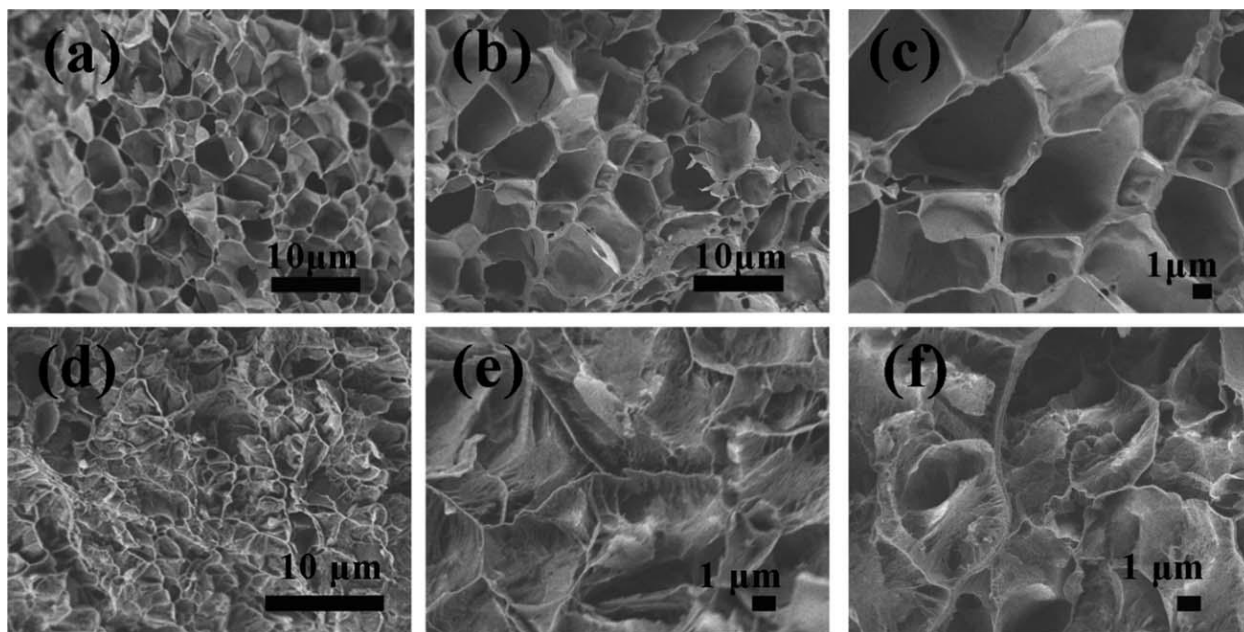
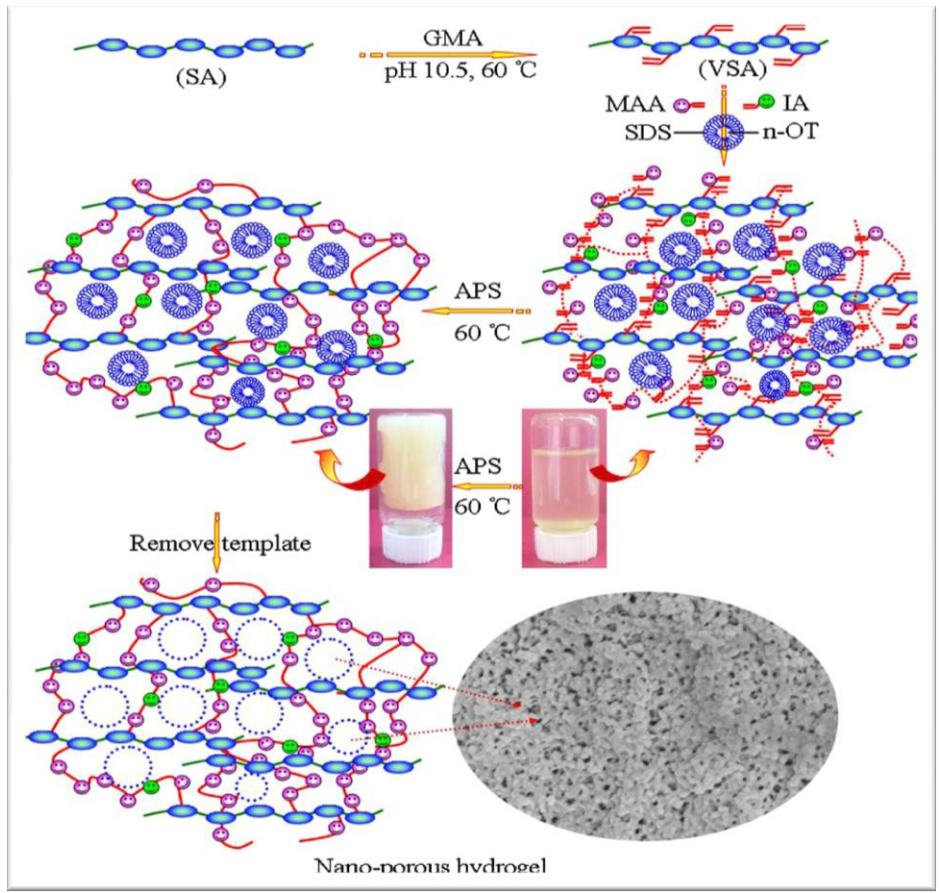


Figure 11. (a) SEM images of (a) pure polyacrylamide hydrogel, (b, c) sodium alginate/polyacrylamide hydrogel, (d-f) graphene oxide/sodium alginate/polyacrylamide ternary hydrogel nano-composite (Fan et al., 2013). Reprinted with permission from (Fan et al., 2013). Copyright 2013 Royal Society of Chemistry.



Figure 12. Different dye solutions before and after the adsorption by graphene oxide/sodium alginate/polyacrylamide ternary hydrogel nano-composite. brilliant green (BG), methylene blue (MB), malachite green (MG), rhodamine 6G (R6G), methyl orange (MO), bordeaux red (BR), calcein (CA) and rose bengal sodium salt (RB) (Fan et al., 2013). Reprinted with permission from (Fan et al., 2013). Copyright 2013 Royal Society of Chemistry.

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**Figure 13. Potential mechanism for the formation of nano-porous vinyl-functionalized sodium alginate-co-methyl acrylic acid-co-itaic acid hydrogel (Wang et al., 2013b).**

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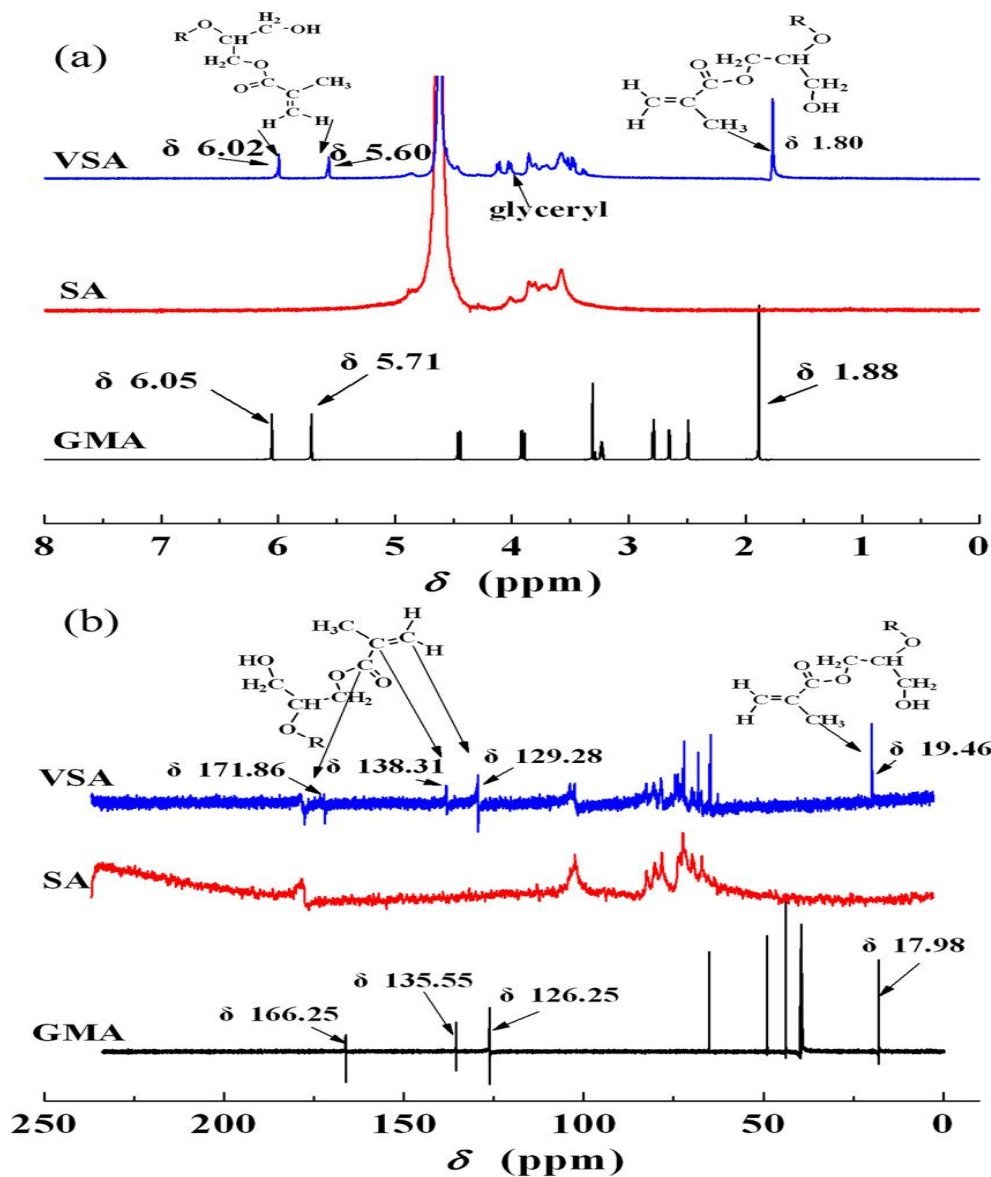
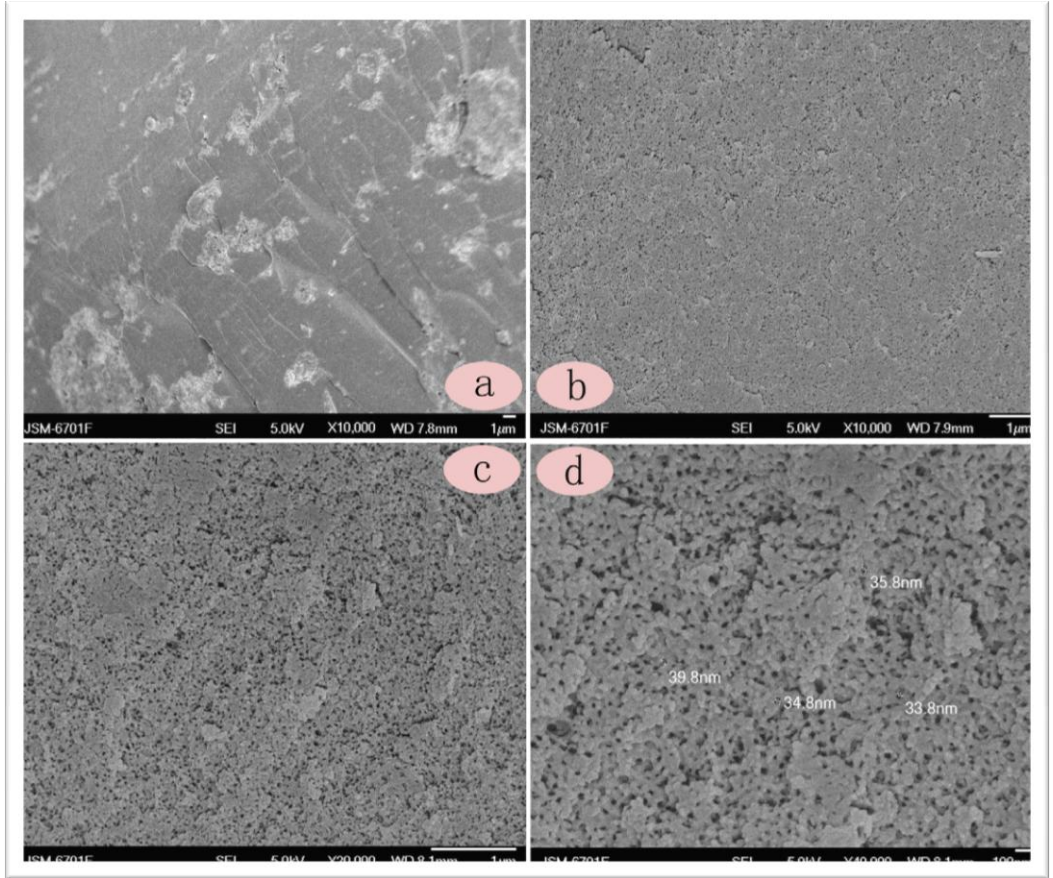


Figure 14. (a)  $^1\text{H}$  NMR spectra of glycidyl methacrylate, sodium alginate and vinyl-functionalized sodium alginate, (b)  $^{13}\text{C}$  NMR spectra of glycidyl methacrylate, sodium alginate and vinyl-functionalized sodium alginate (Wang et al., 2013b). Reprinted with permission from (Wang et al., 2013b). Copyright 2013 Elsevier.

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**Figure 15. FESEM images of (a) non porous vinyl-functionalized sodium alginate-co-methyl acrylic acid-co-italic acid hydrogel, (b-d) nano-porous vinyl-functionalized sodium alginate-co-methyl acrylic acid-co-italic acid hydrogel at different magnification (Wang et al., 2013b). Reprinted with permission from (Wang et al., 2013b). Copyright 2013 Elsevier.**

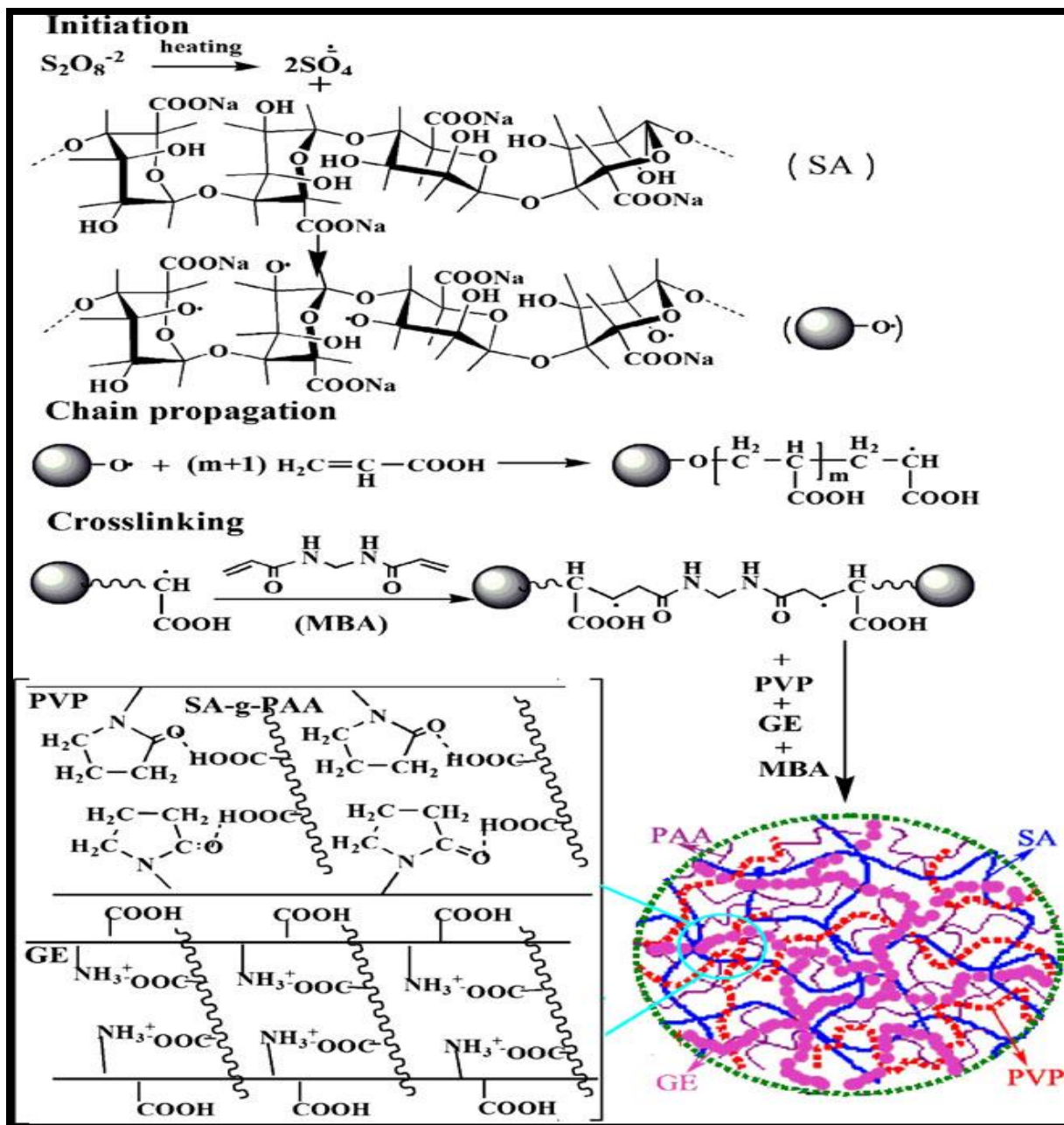
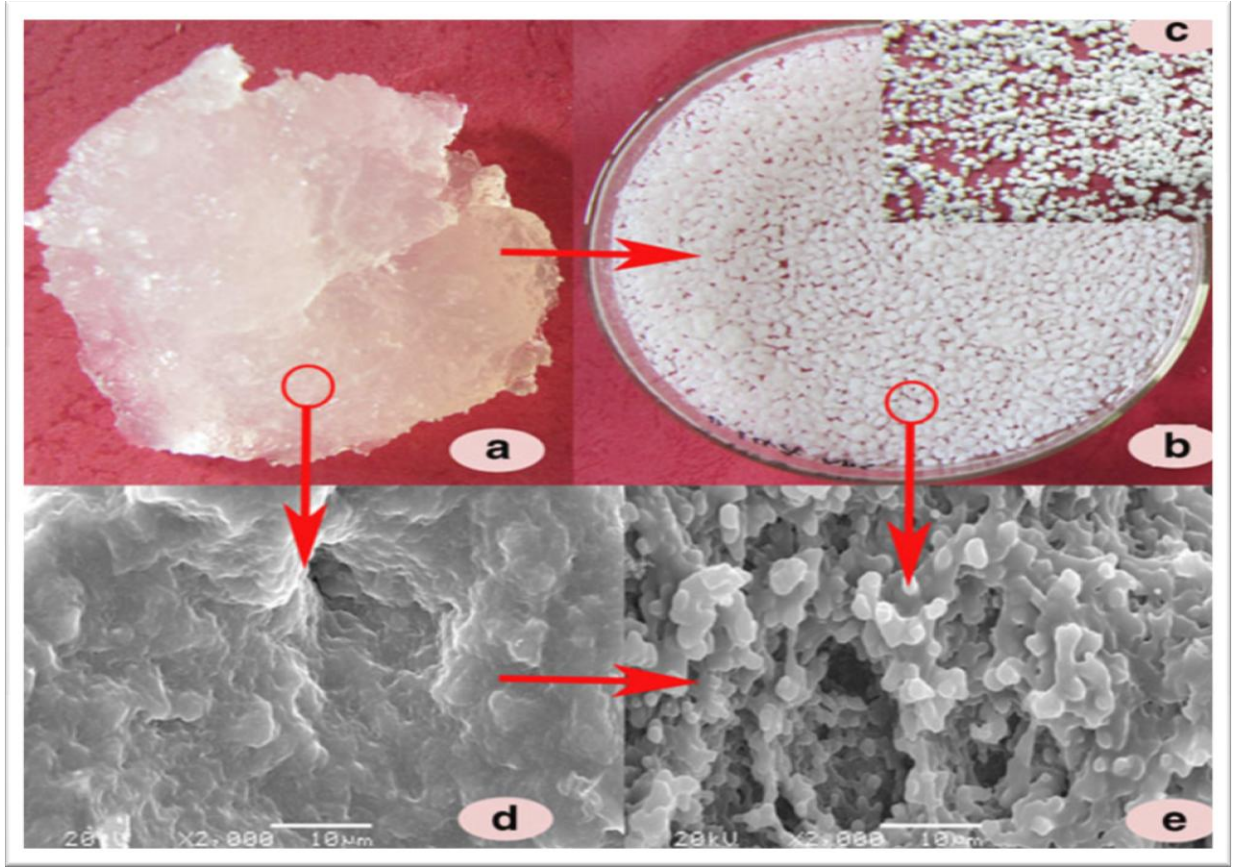


Figure 16. Proposed scheme for the synthesis of sodium alginate-g-polyacrylic acid/polyvinylpyrrolidone/gelatin granular hydrogel (Wang et al., 2013a). Reprinted with permission from (Wang et al., 2013a). Copyright 2013 Springer.

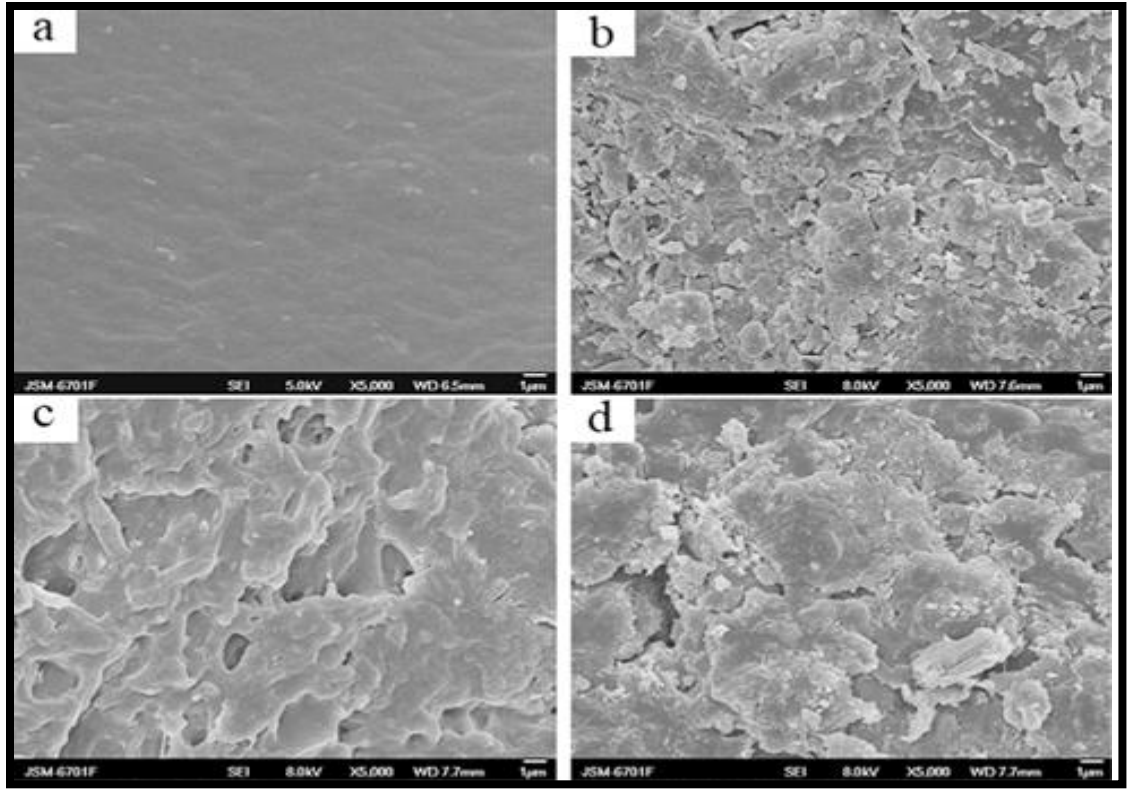


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**Figure 17. Digital images of (a) sodium alginate-g-polyacrylic acid hydrogel, (b) sodium alginate-g-polyacrylic acid/polyvinylpyrrolidone/gelatin granular hydrogel, (c) sodium alginate-g-polyacrylic acid/polyvinylpyrrolidone/gelatin granular hydrogel in dry state. SEM images of (d) sodium alginate-g-polyacrylic acid hydrogel, (e) sodium alginate-g-polyacrylic acid/polyvinylpyrrolidone/gelatin granular hydrogel (Wang et al., 2013a). Reprinted with permission from (Wang et al., 2013a). Copyright 2013 Springer.**

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**Figure 18. FESEM images of (a) sodium alginate-g-sodium acrylate hydrogel, (b) medical stone (c) sodium alginate-g-sodium acrylate/medical stone hydrogel composite with 0.93 g medical stone, (d) sodium alginate-g-sodium acrylate/medical stone hydrogel composite with 3.57 g medical stone (Gao et al., 2011). Reprinted with permission from (Gao et al., 2011). Copyright 2011 Springer.**

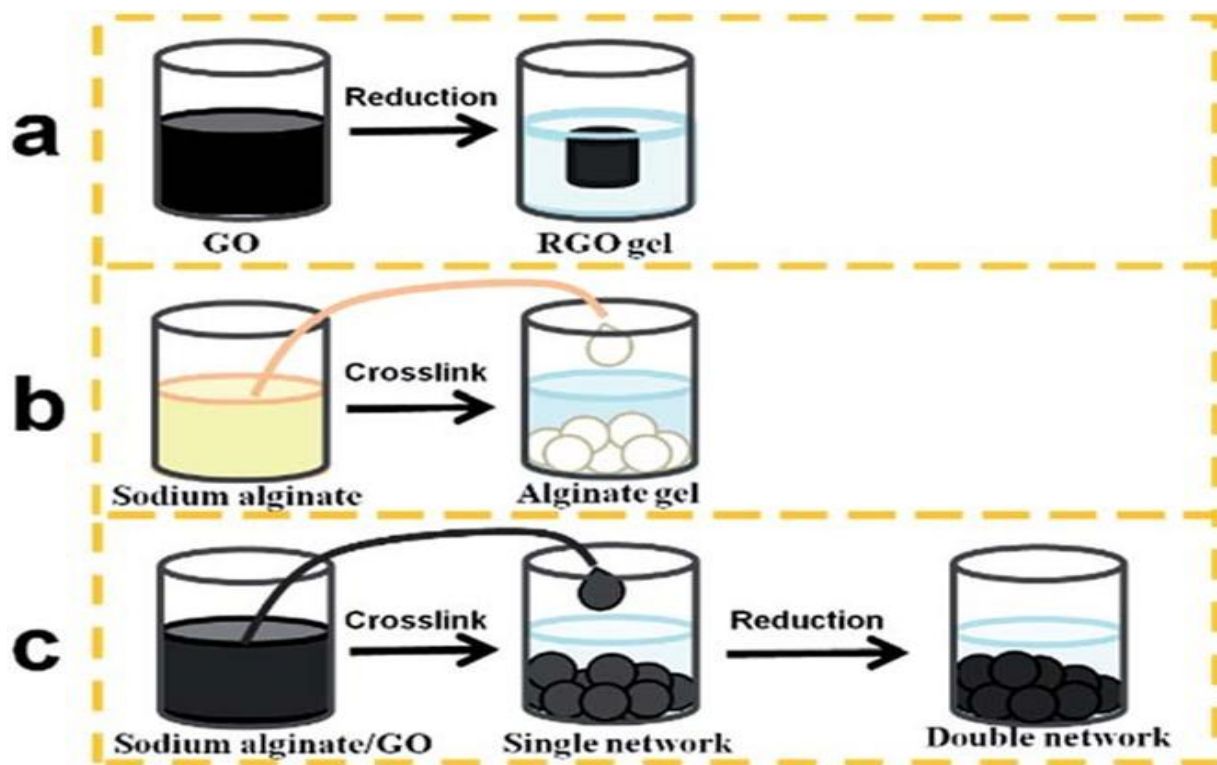
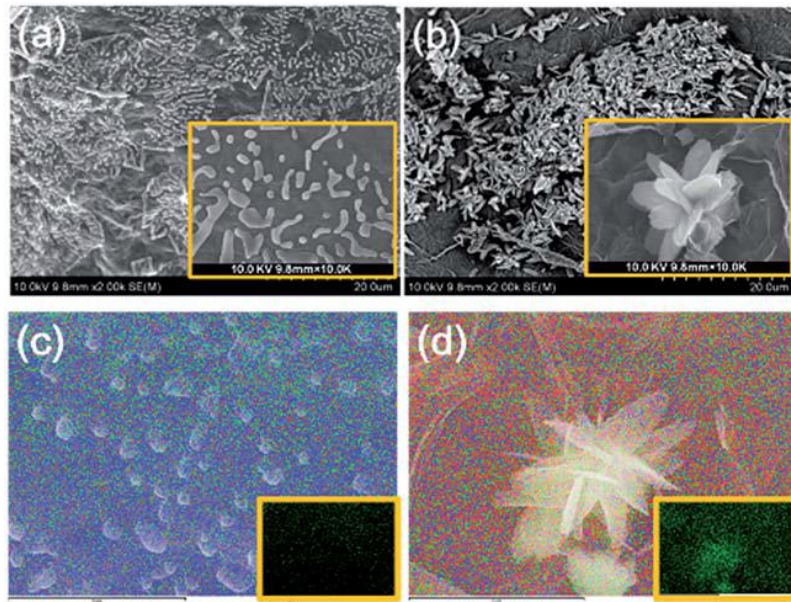


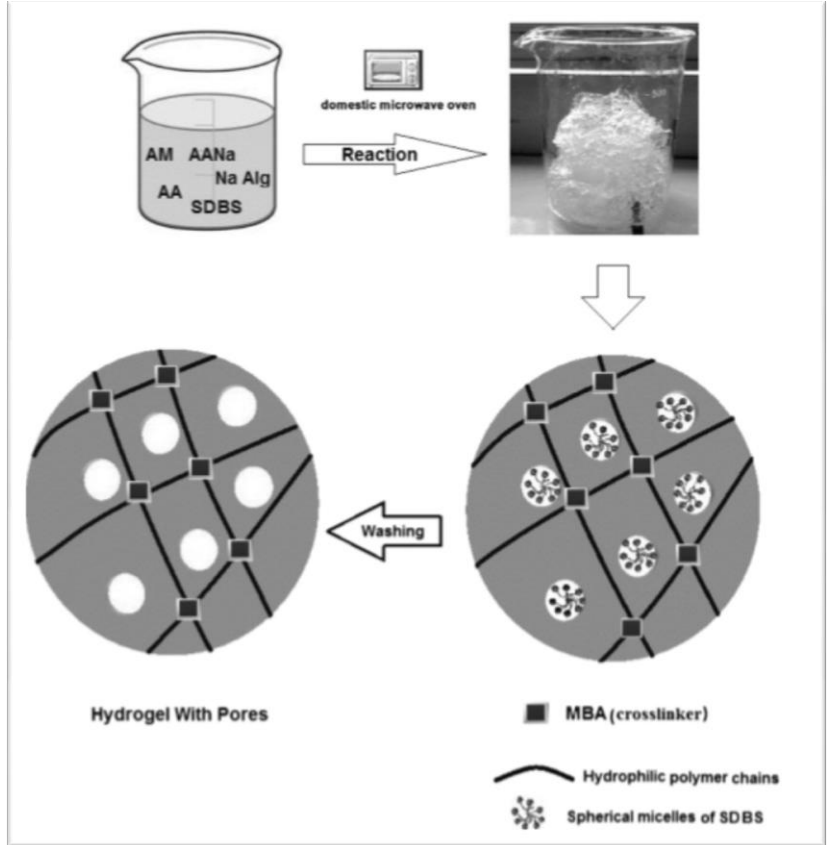
Figure 19. Formation of (a) reduced graphene (RGO) hydrogel, (b) beads of alginate hydrogel, (c) hydrogel bead of graphene/alginate composite (Zhuang et al., 2016).  
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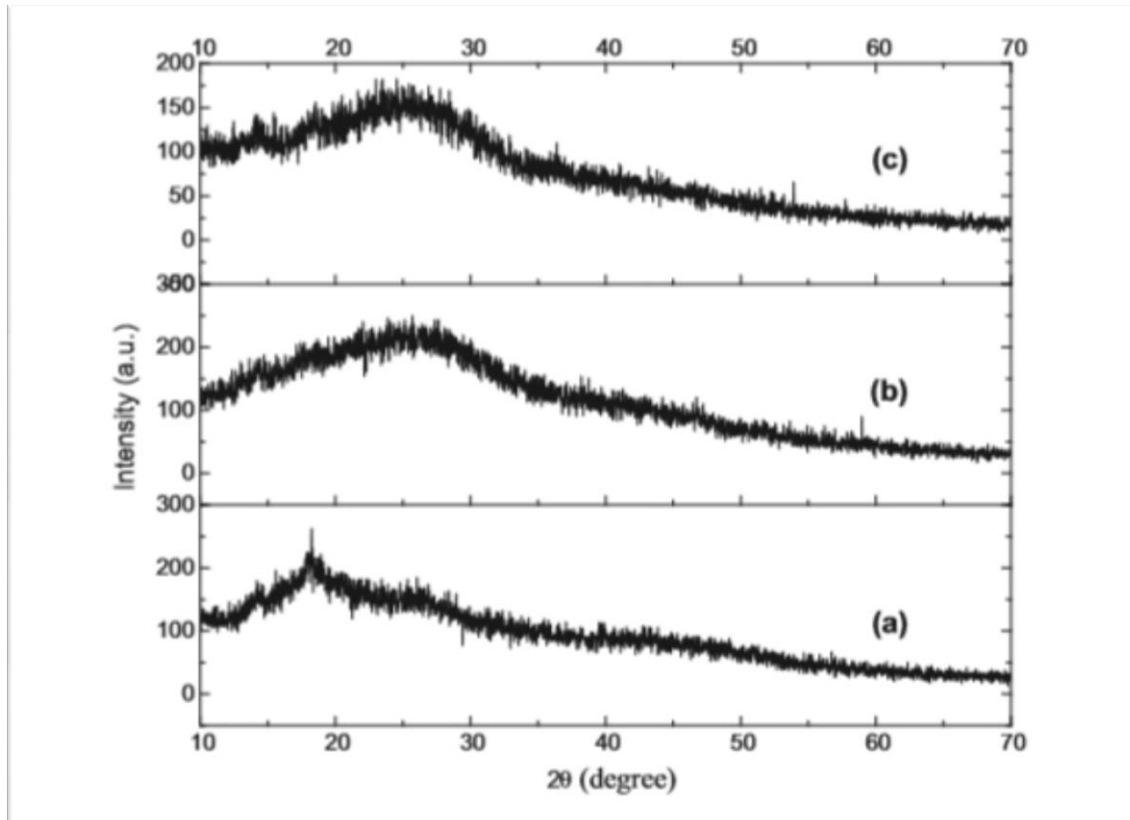
**Figure 20. SEM micrograph of (a) graphene/alginate single network and (b) graphene/alginate double network, elemental mapping of (c) graphene/alginate single network, (d) graphene/alginate double network, with insets of mapping of oxygen element (Zhuang et al., 2016). Reproduced from (Zhuang et al., 2016). Published by Royal Society of Chemistry.**

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**Figure 21. Schematic presentation for the synthesis of sodium alginate-g-poly(acrylic acid-co-acrylamide)hydrogel (Tally and Atassi, 2016). Reprinted with permission from (Tally and Atassi, 2016). Copyright 2016 Springer.**

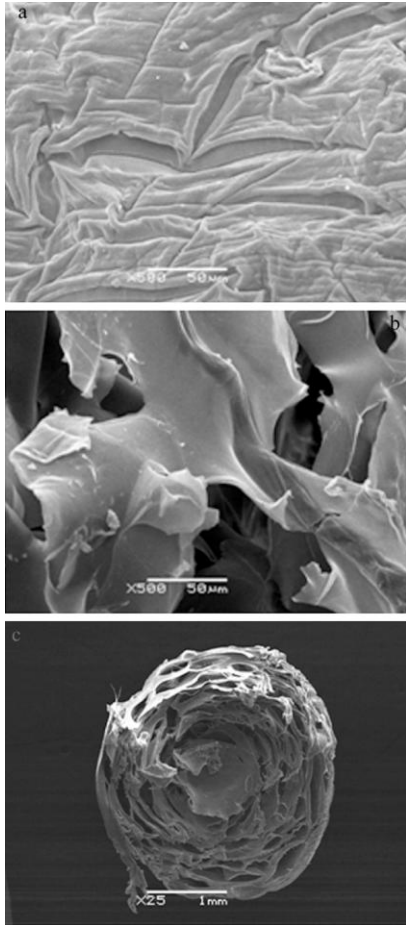




**Figure 22. XRD spectra of (a) sodium alginate, (b) poly(acrylic acid-co-acrylamide), (c) sodium alginate-g-poly(acrylic acid-co-acrylamide) hydrogel(Tally and Atassi, 2016).**

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**Figure 23. SEM images of (a) pure sodium alginate gel bead, (b) sodium alginate-carboxymethyl cellulose gel bead, (c) cross section of sodium alginate-carboxymethyl cellulose gel bead (Ren et al., 2016). Reprinted with permission from (Ren et al., 2016).**

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**Table 1. Sodium alginate based hydrogels for removal of organic pollutants.**

| <b>Sr. No.</b> | <b>Hydrogel Composites</b>  | <b>Organic Pollutants</b>        | <b>References</b>             |
|----------------|---|----------------------------------|-------------------------------|
| 1.             | Sodium alginate–poly(acrylic acid) superabsorbent hydrogel                                  | Victoria blue R and Rhodamine 6G | (Thakur and Arotiba, n.d.)    |
| 2.             | Sodium alginate and silicone dioxide based organic/inorganic nano-composite hydrogel        | Methylene blue                   | (Hosseinzadeh and Abdi, 2017) |
| 3.             | Sodium alginate based titanium dioxide hydrogel beads                                       | Cationic dyes                    | (Lam et al., 2017)            |
| 4.             | Sodium alginate based $\beta$ -cyclodextrin/graphene oxide nano-composite hydrogel          | Methylene blue                   | (Wu et al., 2017)             |
| 5.             | Sodium alginate based TiO <sub>2</sub> nano-composite hydrogel                              | Methyl violet                    | (Thakur and Arotiba, 2017)    |
| 6.             | Sodium alginate/ polyacrylic acid/ TiO <sub>2</sub> nano-composite hydrogel                 | Methylene blue                   | (Thakur et al., 2016)         |
| 7.             | Sodium alginate based silver nano-composite hydrogel  | Methylene blue                   | (Karthiga Devi et al., 2016)  |
| 8.             | Sodium alginate-g-poly (acrylic acid-co-acryl amide)/clinoptilolite nano-composite hydrogel | Methylene blue                   | (Rashidzadeh et al., 2015)    |
| 9.             | Sodium alginate based kappa-carrageenan/sodium montmorillonite nano-composite hydrogel      | Crystal violet                   | (Mahdavinia et al., 2013)     |
| 10.            | Sodium alginate based graphene oxide/polyacrylamide nano-composite hydrogel                 | Cationic and anionic dyes        | (Fan et al., 2013)            |
| 11.            | Sodium alginate based semi-IPN/ acrylic copolymer nano-composite hydrogel                   | Methyl violet                    | (Bhattacharyya and Ray, 2015) |
| 12.            | Sodium alginate based organo-bentonite nano-composite hydrogel                              | Methylene blue and Methyl orange | (Belhouchat et al., 2017)     |

**Table 2. Sodium alginate based hydrogels for removal of inorganic pollutants.**

| <b>Sr. No.</b> | <b>Hydrogel Composites</b>  | <b>Inorganic Pollutant</b>                              | <b>References</b>            |
|----------------|---|---|------------------------------|
| <b>1.</b>      | Sodium alginate based carboxymethyl cellulose hydrogel beads  | Pb(II)  | (Ren et al., 2016)           |
| <b>2.</b>      | Sodium alginate-g-poly (acrylic acid-co-acrylamide) nano-composite hydrogel   | Pb(II), Cd(II), Ni(II) and Cu(II)                       | (Tally and Atassi, 2016)     |
| <b>3.</b>      | Alginate/reduced graphene double-network hydrogel beads and Alginate/reduced graphene single-network hydrogel beads | Cu(II) and Cr <sub>2</sub> O <sub>7</sub> <sup>2-</sup> | (Zhuang et al., 2016)        |
| <b>4.</b>      | Vinyl-functionalized sodium alginate-co-methyl acrylic acid-co-italic acid nano-composite hydrogel                  | Pb(II)  | (Wang et al., 2013b)         |
| <b>5.</b>      | Sodium alginate based granular nano-composite hydrogel  | Ni(II), Cu(II), Zn(II) and Cd(II)                       | (Wang et al., 2013a)         |
| <b>6.</b>      | Sodium alginate-g-poly (sodium acrylate)/medical stone nano-composite hydrogel                                      | Ni(II), Cu(II), Zn(II) and Cd(II)                       | (Gao et al., 2011)           |
| <b>7.</b>      | Tetra sodium thiacalix [4] arenetetrasulfonate-sorption-sodium alginate nano-composite hydrogel                     | Ni(II), Cu(II), Pb(II), Cd(II), Co(II) and Cr(III)      | (Lakouraj et al., 2014)      |
| <b>8.</b>      | Sodium alginate hydrogel beads by post cross-linking  | Ag(I), Cu(II) and Fe(III)                               | (Lu et al., 2015)            |
| <b>9.</b>      | Sodium alginate based membrane - filtration   | Ca(II) and Mg(II)                                       | (Fatin-Rouge et al., 2006)   |
| <b>10.</b>     | Chitosan-alginate hydrogel beads and chitosan-glycolic acid hydrogel beads  | Cu(II)  | (Nghah and Fatinathan, 2008) |

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