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Recent Review of Hydrogels as favourable Adsorbent of Heavy Metal Ions for Effluent Water Treatment.

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Abstract
The pollution of water sources has become right into worldwide hassle because of the indiscriminate disposal of pollutants both organic and inorganic in nature Industrial growth meets our needs and promotes economic development. However, pollutants from such industries pollute water bodies posing a high risk to organisms. Researchers were therefore asked to develop efficient methods to remove toxic heavy metal ions from water bodies. The adsorption process has shown promising results in the removal of heavy metal ions and is easy to operate at large scale, making it applicable for practical applications. Numerous sorbents, including hydrogels, have been developed and reported and have received great attention due to their reusability, ease of fabrication, and handling. Hydrogels are generally made by cross-linking polymers, resulting in a three-dimensional structure that exhibits high porosity and high functionality. Due to their functional groups, they are naturally hydrophilic and non-toxic. Therefore, this review article provides different methods for preparing hydrogel sorbents and summarizes recent progress in removing heavy metal ions using hydrogel sorbents. In addition, we briefly discuss the mechanisms involved in the removal of heavy metal ions. A detailed review of these factors affecting the interaction between hydrogels and heavy metal ions are stated which are used for the adsorption of heavy metal ions.

Keywords: *Hydrogels, Heavy metal ions, Waste Water Treatment, Adsorption.*

I. Introduction:

The main sources of heavy metal ions are pesticides, fungicides, refineries, fertilizers, mining, smoking, fission plants, chemical industry, paints, electroplating, welding, automobiles, batteries, etc. [1]. Heavy metal ion contamination damages water systems and human organs such as lungs, kidneys, central nervous system, nose, skin, gastrointestinal tract and brain.[2]

The presence of heavy metals in wastewater is increasing with the growth of industry and human activities. Applications in metal smelting, petrochemicals, papermaking and electrolysis. Wastewater contaminated with heavy metals is released into the environment, threatening human health and ecosystems. Because heavy metals are non-biodegradable and carcinogenic, the presence of these metals in water in inappropriate amounts can pose serious health problems for living organisms [3].

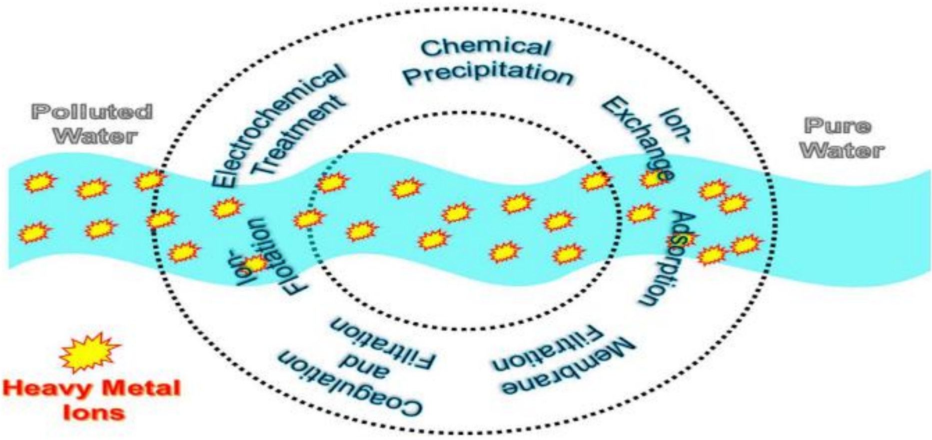

Fig-01 Various Methods for Heavy metal ion removal.
[Source Online https://www.mdpi.com/metals /html/images/metals-11-00864-g002.png]

The metals such as Silver (Ag), Iron (Fe), Manganese (Mn), Molybdenum (Mo), Boron (B), Calcium (Ca), Antimony (Sb), Cobalt (Co), etc. are commonly available in wastewater hence need to be removed [4].
Many efforts have been put forward to remove the heavy metal ions from polluted water. .Fig 1 illustrates the different methods to purify the polluted water such as chemical precipitation [5], coagulation and flocculation [6], membrane filtration [7], ion flotation [8], ion-exchange [9] electrochemical treatment [10], and adsorption [11].

Amongst all these adsorption remains the simplest and cheapest approach to remove contaminants from wastewater, and even naturally occurring sorbent materials have received a great deal of attention.[12] Promising examples of such sorbents include: Hydrogel (HG), which represents a three-dimensional polymer network of hydrophilic groups with high capacity to adsorb numerous metal ions and dyes from wastewater. Growing interest in the development and application of novel hydrogels in wastewater treatment has been attributed to their precise chemical properties in addition to their hydrophilicity, sensitivity, and functionality. Hydrogels have excellent overall performance in the adsorptive removal of various aqueous contaminants in addition to heavy metals, nutrients and toxic dyes. The design of adsorbent hydrogels leads to the inheritance of the advantages of each component and offers new features from the synergistic effects between the components, thus opening new application areas [13].

II. Review of Literature

Researchers around the world are concerned about the effects of increasing sewage pollution on the atmosphere.. Therefore, wastewater treatment is still essential before it enters the natural water stream. Removal of heavy metal ions by hydrogels depends on many factors, including: Experimental conditions and reaction sites available in hydrogels. These factors influence the interaction between hydrogel and heavy metal ions. Adsorption experiments have been reported for single and multiple systems. Some hydrogels used to adsorb heavy metal ions are regenerated and used again and again [14].

Hydrogels can be positioned as multifunctional platforms for deep purification of micro contaminated water and specific detection of microcontaminants. Various scientist have synthesized hydrogels and studied its adsorption on waste water treatment especially with removal of heavy metal ions. Many researchers including Li et al., and Tamas et al., [15-16], have stated in their review that adsorption is commonly recognized as a potential method to remove various pollutants.

Yang et al., [17] worked on the Polyvinyl alcohol (PVA) which is a widely used non-toxic, biodegradable, water-soluble linear polymer, which has the inherent advantages of preparing ideal "green" materials. Sahoo et al., [18], further emphasized on that the polymer material prepared with PVA is not only non-toxic, but also has good biocompatibility, good viscoelasticity, strength and processability.

Choi, J.-W.; Kim, H.J and team [19], synthesized a higher capacity hydrogel along with a grapheme oxide, alignate and polynitrile with a moderately cross-linked network structure, which contains abundant functional groups in its unique network . These functional groups in the hydrogel's three-dimensional network adsorbed Cupper metal ions into the network through hydrogen bonds, electrostatic force and chemical complex interaction, and the network space accommodated these adsorbed ions, so as to achieve high-capacity adsorption in aqueous layer. Yung Xu et al [20], in the review described the hemicellulose based hydrogels with superior applications as adsorbents with higher adsorption capacity, faster adsorption rate and excellent desorption-regeneration ability are commonly regarded as highly efficient adsorbents. Kaur et al., [21], in the review stated that with the increasing attention on the environmental friendliness of materials, people are more inclined to use natural polymers to synthesize hydrogel materials for various purposes, because natural polymers have the advantages of being renewable, cheap, non-toxic and environmentally friendly.

Nan Hou et al [22], Fabricated Hydrogels via Host−Guest Polymers as Highly Efficient Organic Dye Adsorbents for Wastewater Treatment New self-assembled hydrogel materials of poly(vinyl alcohol)/cyclodextrin-modified poly(acrylic acid)/azobenzene-modified poly(acrylic acid) (PVA/PAA-CD/PAA-Azo) were successfully prepared via host-guest interactions and hydrogen bonds.

Vakili et al [23], presented, a variety of innovative technique that has been developed and employed to decontaminate the harmful Pb(II) ions from polluted water In their research findings Ezeokonkwo et al., and Alyafei A et al., [24-25], concluded that using adsorption materials to remove heavy metal pollutants in water through physical or chemical action is considered to be one of the effective ways to eliminate Pb(II) pollution. The design and development of new adsorbents with high adsorption capacity and fast adsorption rate have received considerable retention in both academic and industrial domains.

Hashem et al.,[26], by combining the two cheap materials, CS and HNTs, fabricated novel hydrogel beads that can be applied for removal of MB and malachite green (MG) from wastewater. CS–HNTs hydrogel beads were obtained by the dropping and pH-precipitation method.

Hoang et al.,[27], concluded that Poly(N-vinyl-2-pyrrolidone) (PVP) is a water-soluble polymer that is quite friendly to the environment and to humans due to its non-toxic, biodegradable, and biocompatible properties. Owing to these advantages, PVP is totally suitable for synthesize hydrogel materials. Hydrogels based on the interaction of PVP and other component for removing anionic dyes and metal ions.

Maria Demetal and Co Workers [28] in their work, highly elastic and superabsorbent hydrogels of collagen-poly(vinyl pyrrolidone) (PVP)-poly(acrylic acid) (PAA)-poly(ethylene oxide) (PEO) quaternary copolymer were obtained using e-beam crosslinking reaction to be used for soft tissue engineering. This study is focused on the evaluation of rheological, swelling and stability properties under different pH media, offering in the same time an insight on the hydrogel network parameters. Rheological studies confirm that collagen/PVP/PAA/PEO hydrogels present solid behaviour similar with that of most soft tissue as human abdominal fat, skin and dermis and even more of breast gland or brain and liver. Hydrogels revealed superabsorbent capacity, both in deionized water and in simulated biological buffers, 1600% and 4000%. The stability and physical form of hydrogels were maintained for more than 50 h in environments that simulate both the pH of the healthy skin and that of an infected wound. The elastic modulus and the parameters of the network structure of hydrogels showed the formation of hydrogel with permanent network.

Chelating poly(vinylpyrrolidone/acrylic acid) (PVP/AAc) copolymer hydrogels were prepared by radiation-induced copolymerization by El Hag Ali and Co Workers[29]. The effects of preparation parameters such as PVP content in the hydrogel and irradiation dose on the swelling behaviour of the hydrogel were studied. The pH dependent swelling was investigated. The thermal stability of the prepared hydrogel and the metal chelated ones was characterized by TGA. The removal of Fe(III), Cu(II), and Mn(II) from aqueous solution by the prepared PVP/AAc chelating hydrogel was examined by batch equilibration technique. The influence of treatment time, pH, and the initial feed concentration on the amount of the metal ions removed was studied. The results show that the removal of the metal ion followed the following order: Fe(III) > Cu(II) > Mn(II). The amounts of the removed metal ions increased with treatment time and pH of the medium.

Yosthanase Tassanapukdee and co workers [30] as illustrated in fig 02 simple protocol of chitosan/polyvinyl alcohol/polyvinylpyrrolidone (CS/PVA/PVP) hydrogel synthesis using microwave-assisted irradiation to initiate and accelerate network formation was explored.

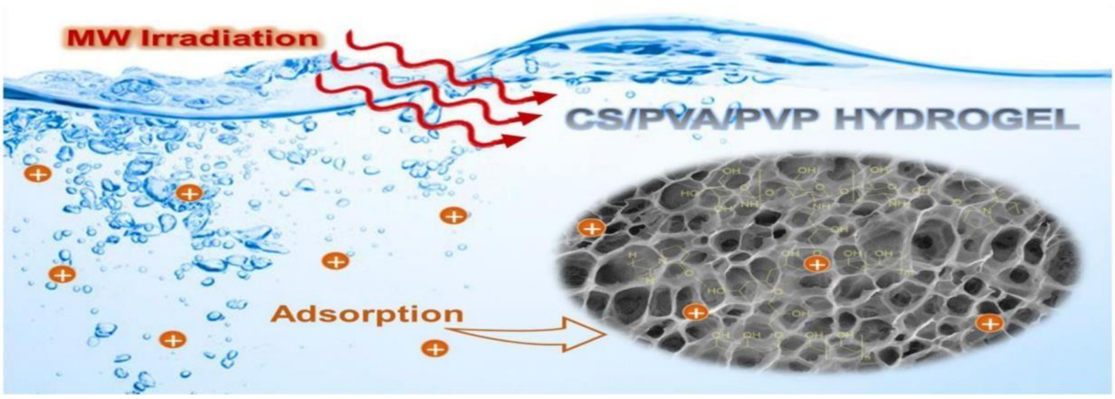

Fig.02 CS/ PVA/ PVP hydrogel [30].

The optimum ratio of CS:PVA:PVP was 0.3:0.6:0.3 g with epichlorohydrin as a cross-linking agent under 600 watts of irradiation for 3 min. The gel swelling degree and gel fraction of the hydrogel were approximately 1627.4% and 42.6%, respectively. The desorption and reusability study of the CS/PVA/PVP hydrogel revealed that 0.1 M EDTA was a suitable candidate to regenerate the hydrogel as more than 60% of metal ion removal efficiency was still achieved after five adsorption cycles.

Luqman shah et.al [31] reports an efficient removal of selected heavy metal ions using a low- cost superabsorbent polymer hydrogel (SPH) composed of acrylic acid and acrylamide in different compositions which were prepared by single step free radical polymerization technique using ammonium persulphate and N, N-methylene bis-acrylamide as an initiator and cross-linker respectively. All the samples were highly effective in the removal of Cd^{2+}, Ni^{2+}, Cu^{2+} and Co^{2+} from aqueous medium at pH range of 2–10 following pseudo second order kinetics and Freundlich adsorption model. Also, the removal capacity was greater at pH 7 and materials showed high selectivity towards Co^{2+} and Cu^{2+} in competitive removal process. The high removal ability >75% for each metal ion, make these materials as an efficient, easily obtainable, and environmental friendly product.

Another eco-friendly way of waste water treatment was done by the use of cotton fabrics. Massive consumption of cotton fabrics has brought up a serious problem concerning the waste cotton fabrics (WCFs) disposal. It is widely accepted that if WCFs can be reutilized, there will be great business potentials. Jainhu ma and Co workers [32] prepared a double network hydrogel based on WCFs and polyacrylamide (Cellulose/PAM

DNHs) for heavy metal removal. The DNHs exhibit fast kinetics that sorption equilibrium is achieved in 5 min because of the porous and sheet-like laminar structures they possess. The DNHs also illustrate excellent adsorption property and good reusability.This work provides a new avenue for the combination of WCFs reuse and heavy metal removal, which is of great importance to the construction of resource sustainability and environment-friendly society.

A novel poly(Hydroxyethyl methacrylate/Maleamic acid) (p(HEA/MALA)) hydrogel was synthesized by Ningmei Wu and Zeingkei Lee[33] ^{60}Co-γ induced copolymerization, and used to remove Pb^{2+}, Cd^{2+}, Ni^{2+} and Cu^{2+} from aqueous solutions. FTIR spectra and TGA analysis showed that the p(HEA/MALA) hydrogel was indeed a copolymer of HEA and Maleamic acid. Adsorption kinetics of Pb^{2+}, Cd^{2+}, Ni^{2+} and Cu^{2+} ions on p(HEA/MALA) followed pseudo-second-order kinetic model, and the adsorption rates followed the order $Cd^{2+} > Pb^{2+} > Ni^{2+} > Cu^{2+}$. According to the competitive adsorption results, the priority order in multi-component adsorption was $Pb^{2+} > Cu^{2+} > Ni^{2+} > Cd^{2+}$. These findings suggest that the hydrogel is a promising adsorbent to separate and recover the heavy metal ions from contaminated water.

Noman Chowdhury et al [34], have successfully synthesized different ionogenic hydrogels via a simple free radical polymerization method. AAm is used as a main monomer; one or more from AAc, AMPS, and MEDSA are used as comonomers to make poly(AAm-AAc), poly(AAm-AMPS), poly(AAm-AAc-AMPS), poly(AAm-MEDSA) hydrogels with varying monomer and comonomer concentrations. The hydrogels exhibit temperature and pH sensitivity. The reported hydrogels can remove heavy metal ions (Fe^{3+}, Hg^{2+}, and Cr^{3+}) from aqueous samples.

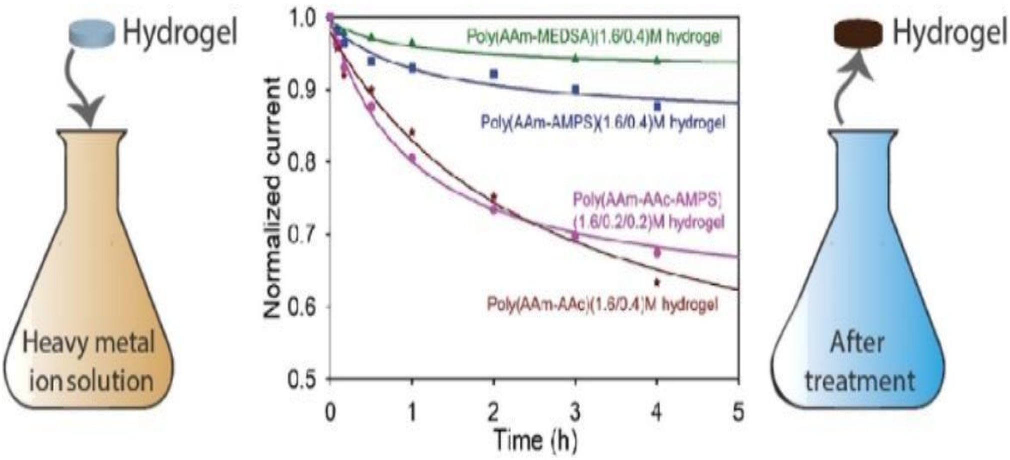

Fig 03 graphical representation of different parameters of Aam Monomer [34]

The enhanced compressive strength and thermal stability of hydrogels with the incorporation of metal ions will expand their area of applications and open up windows to further investigate the mechanical property improvement of conventional hydrogels using metal ions as an additional cross-linker. The synthesized hydrogels can possibly be used for the treatment of industrial wastewater, water purification, and so on.

Recently Chenxi Zhao et al[35] prepared Polysaccharide-based biopolymer hydrogels to expand their performance and usage, chemical modification of single polysaccharide hydrogels was carried out to introduce functionalized groups, and the final composite hydrogels obtained was greatly enhanced in mechanical strength and adsorption properties, in addition to making up for some defects. The composite polysaccharide-based hydrogel, has an efficient adsorbent, can adsorb heavy metal ions rapidly and in large quantities, and was easily recoverable and reusable. Thus polysaccharide-based hydrogels with more stability, specificity and adsorption capacity have been developed for wider applications in water treatment.

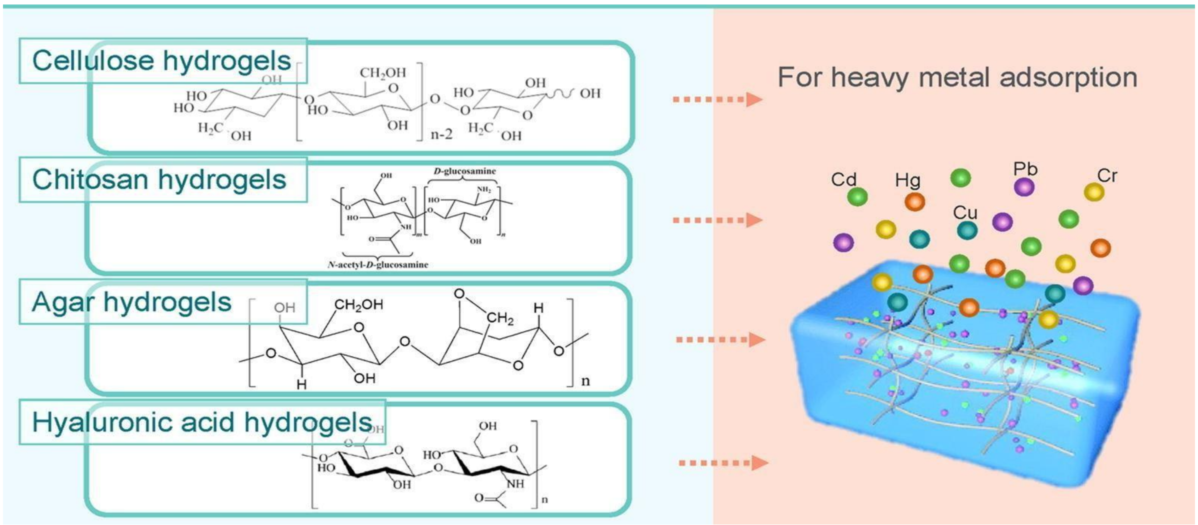

Fig 03 Polysaccharide based hydrogels for heavy metal ion adsorption.(Web of Science)

III. Conclusion

Contamination of water bodies by heavy metal ions requires an urgent response. Among existing processes adsorption is a simple, effective and inexpensive method. Review of various methods for making hydrogel sorbents was presented in this review. Selected studies on the modification of sorbents for efficient removal of heavy metal ions and the plausible interactions between sorbents and heavy metal ions are also briefly described. In this most widely applicable hydrogels were discussed as effective adsorbents for heavy metal ions. However, some important aspects should be considered when developing hydrogel sorbents for heavy metal ions. Hydrogels are recycled and reused for many cycles. However, the reported results show a decrease in removal efficiency with cycling. Therefore, research should focus on developing hydrogel sorbents that maintain removal efficiency. In addition, hydrogels tend to remove one or several metal ions, and it is difficult to completely remove all metal ions. Therefore, additional research should be focused on developing effective sorbents for simultaneous and complete removal of multiple heavy metal ions. This enables the practical use of adsorbents to remove multiple heavy metal ions.

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