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An Overview on Activated Carbon and Zeolites in Water Treatment

Steffin John Eapen[1], Dilson B .Samuel[2] & Manikandan P.M[3]
[1,2]Dept. of Chemical engineering, TKM College of Engineering, Kollam
[1]steffin121@gmail.com, [2]dilsonbsamuel.db@gmail.com, [3]mpmmanikandan@gmail.com

***Abstract**— Water treatment has become essential so as to increase water supply for domestic purposes. At present, there are different kinds of methods by which water or effluent water can be treated. Among these, adsorption stands out as an effective and better option. Activated Carbon and Zeolites are commonly used adsorbents. Under zeolites, we have natural zeolites, zeolites made from fly ash and also surfactant modified zeolites. All of these had different properties at different conditions. But Activated Carbon or zeolites on its own are selectively adsorbing. Therefore, there is a need to synthesis a blend of the two in a particular composition. This paper reviews and evaluates from literature to how selective characteristics of activated carbon and zeolites can be combined for better composites.*

***Index Terms**—Activated Carbon, Surfactants, Water treatment, Zeolites*

I. INTRODUCTION

Surfactants play a major role in our day to day life. Major types of surfactants like soaps and detergents etc. are useful when they come in contact with water. Hence, studies conducted on surfactants modified with activated carbon and zeolites are beneficiary as they can be used to treat contaminated water as well. The world requires large quantities of domestic water to fulfill its domestic or day today needs. However, most of it gets contaminated by the untreated disposal of wastes from industries which results in the release of ions of heavy metals such as cadmium, lead, zinc, cobalt etc. These impurities exists in multiple forms such as microorganisms, suspended solids or dissolved solids or colloidal inorganic and organic substances [1]. Water treatment can be carried out in many ways such as physical, chemical and biological process. Suitable examples would be microbial degradation, filtration, chemical oxidation, electrochemical oxidation, coagulation and membrane separation [2], [3]. However, these techniques have their own limitations such as high cost, generation of secondary pollutants and poor removal efficiency [2], [3].But, adsorption is efficiently using technique in effluent water treatment. The benefit of adsorbents are superior in terms of initial cost, secondary harmful substances and ease of operation. Biodegradation suffers from optimization problems while ion exchange and chemical oxidation are considered to be high-cost methods. Electrochemical processes suffer from three main problems: [1] high cost and unstable anodic materials; [2] the effluent cannot be discharged in the presence of high chloride concentration; [3] low columbic efficiency. Furthermore, floatation coagulation methods have a low efficiency. More sophisticated techniques like solvent extraction, bio-sorption, and ultra-filtration are either expensive or cannot cope with high concentrations of contaminants. All these methods have significant disadvantage such as incomplete ion removal, high energy requirements, and production of toxic sludge or other waste products that require further disposal [4]. Among different adsorbents used, activated carbon is used for a long time for this purpose [5], [6], [7]. But its high cost of production, generation of pollutants and poor removal efficiency prove a hindrance for adsorption [7], [8].Studies have shown that natural zeolite, being an abundant resource that is available globally, can be substituted for activated carbon because of its high cation-exchange capacity and surface area, etc. The above mentioned substituents such as Activated Carbon, natural zeolites, surfactant modified zeolites are being used for effluent water treatment.

II. ACTIVATED CARBON

Activated carbons (AC) are known as very effective adsorbents due to their highly developed porosity, large surface area, variable characteristics of surface chemistry, and high degree of surface reactivity [37]. They are used in water treatment as catalysts and catalyst supports used for different purposes such as the removal of pollutants from gaseous or liquid phases and the purification or recovery of chemicals [38]. The treatment of wastewater and contaminated groundwater using AC is increasing throughout the world as a result of the limited sources of water supply [39].In such treatments, AC is normally used as a primary treatment, preceding other purification processes, or as a final tertiary or advanced treatment. When using AC, the adsorption process results from interactions between the carbon surface and the adsorbate. These interactions can be electrostatic or non-electrostatic. When the adsorbate is an electrolyte that dissociates in aqueous solution, electrostatic interactions occur; the nature of these interactions, that can be attractive or repulsive, depends on the: (i) charge density of the carbon surface; (ii) chemical characteristics of the adsorbate; and (iii) ionic strength of the solution. Non-electrostatic interactions are always attractive and can include: (i) van der Waals forces; (ii) hydrophobic interactions; and (iii) hydrogen bonding. As(V) and As(III) removal from water was studied using a char carbon (CC) derived from fly ash. CC and AC adsorbents removed equal amounts of As(V) at optimum condition. However, percent As(III) removal was more on CC than AC [40].As(V) removal by an iron-oxide impregnated activated carbon was modeled by Vaughan and Reed.[41]. Several low-cost adsorbents have been tested; however, AC are known to have much better performances to treat contaminated effluents. The need for efficient and economic removal of pollutants, namely from aqueous phase, resulted in the development of research on the use of waste materials as precursors for the preparation of less costly AC. The use of such materials can be, therefore, an efficient alternative for both, production of low-cost AC, and adoption of effective waste management practices [42].

III. NATURAL ZEOLITES

Water treatment using natural zeolites are carried out mainly by ion-exchange method by which the dissolved cations present in water are removed by zeolite exchange site. Study by Denes Kallo concluded that the most common cation in waters affecting human and animal health is $NH_4^{+.}$ It can be removed by exchanging with biologically acceptable cations such as Na^+, K^+, $Mg^{2+,}$ Ca^{2+} or H^+ residing on the exchange sites of the zeolite. Fortunately, many natural zeolites (e.g. clinoptilolite, mordenite, phillipsite, chabazite) are selective for NH^{4+} (vide infra), meaning that they will exchange NH^{4+} even in the presence of larger amounts of competing cations. Clinoptilolite and mordenite are also selective for transition metals (e.g. Cu^{2+}, Ag^+, Zn^{2+}, $Cd^{2+,}$ Hg^{2+}, Pb^{2+}, Cr^{3+}, Mo^{2+}, Mn^{2+}, Co^{2+}, Ni^{2+}), which are often present in industrial waters and can be very toxic even in concentration as low as several mg/L. As emphasized in discussions of radioactive waste treatments, both linoptilolite and mordenite have very high selectivities for Cs^+ and Sr^{2+} and can therefore be used to remove minute amounts of radioactive ^{137}Cs and ^{90}Sr from nuclear process wastewaters. Natural zeolites are primarily used for removing organic impurities such as hydrocarbons, oxygen-containing compounds, halogenated derivatives, amines, humic acids, proteins, and lipids. Most organic molecules or particles are too large to penetrate into channels and cages to access the extra framework exchange sites of natural zeolites [1].

Their adsorption take place on the surfaces of the zeolite containing materials (e.g. external crystal surfaces of zeolites), which can have a surface area as large as several 10 m2/g. Volumetrically, most important natural zeolites on the surface of the Earth are of sedimentary origin, and zeolites in such deposits occur in clusters of crystals often having inter crystalline pore sizes of 10 to 1000 nm in diameter (i.e. rock pores). Colloids, enzymes or microorganisms as large as bacteria can be trapped within these intra-particle pores. As a result of the large surface area, which is accessible for adhering bacteria, natural zeolites can become effective bio-filters when compared with particles having smaller total surface areas such as quartz sand beds[9].

REMOVAL OF MICROORGANISMS

One of natural zeolite like clinoptilolite used for removal of NH_4^+ions and H_2S from waste water [10]. A variety of microorganisms, such as *Escherichia coli*, poliovirus, *coxackie* virus, and bacteriophages, have been effectively removed from drinking water using an Al_2 $(SO_4)_3$ coagulant and clinoptilolite-rich material [11]. The numbers of microorganisms decreased by 50% when clinoptilolite-rich material or coagulant alone was used to treat the water, whereas the two additives together removed 90% of all microorganisms during the same time period. This is due to the strong bond between microorganism-clinoptilolite-coagulate. An apparatus for removal of NH^{4+}, Fe^{2+}, Mn^{2+} etc. from drinking water by clinoptilolite materials has been disclosed in a Hungarian Patent and built in Hungary [12]. Another natural zeolite phillipsite was used to remove the indicator bacteria such as *coliforms*, fecal *coliforms*, fecal *streptococci*, and dissolved organic matter [11].

REMOVAL OF HEAVY METALS

Studies have shown that natural zeolite can effectively remove the suspended and dissolved particles from effluent water from municipal and agricultural waste water and zeolite treatments using Natural zeolite has proved to be superior to conventional biological waste water treatment [1]. It is also used for remediation of waste water containing heavy metals cations like Cu^{2+}, Cd^{2+}, Zn^{2+}, Ni^{2+} and Pb^{2+} and radioactive cations as well. As a result of the low cost of natural zeolites and the fact that their native exchangeable cations are relatively safe to humans, plants, and animals (e.g. Na+, Ca^{2+}, Mg^{2+}, K^+), natural zeolites are especially attractive alternatives for removing undesirable heavy-metal ions from effluent wastewaters mainly of industrial origin [16]. Study conducted by Ames (1959) concluded that clinoptilolite-rich tuff , was the most promising of 15 different zeolites tested for Cs^+ removal from solutions containing competing cations such as Al^{3+}, Fe^{3+}, Ba^{2+}, Sr^{2+}, Ca^{2+}, Mg^{2+}, Rb^+, K^+, NH^{4+}, Na^+, and Li^+ in large concentrations[13]. The advantage of natural zeolites over organic ion-exchange resins lies in their resistance to degradation in the presence of ionizing radiation and their low solubility. Zeolites can also be used for long-term storage of long-lived radioisotopes and further, Mercer and Ames (1978) presented a detailed description of the use of natural zeolites for the removal and fixation of radionuclides, including removal of ^{137}Cs from high-level radioactive waste, decontamination of low- and intermediate-level wastes and fixation of radioactive wastes for long-term storage. They found that other natural zeolites, including chabazite, erionite, and mordenite, in addition to clinoptilolite, have high selectivities for Cs^+ [14], [15] .Mn^{2+} has been removed from drinking water using simple ion exchange with natural clinoptilolite-rich zeolite[16] . Also it has been added directly to sewage sludge in attempts to reduce the release of heavy metals when the treated sludge is applied as a beneficial source of organic matter and plant nutrients [17]. Studies conducted by Pabalan and Bertetti shown that ion-exchange selectivities of zeolites depended on the ion concentration of the metals in the wastewater to be treated as well as cation-exchange selectivities for most transition or heavy-metal cations became higher compared with alkali or alkaline-earth cations by lowering the concentrations of transition and heavy-metal cations in water bodies.[17]. Natural zeolites play a major role in in cleaning or remediating industrial effluent water which are often laden with heavy metals due to increasing salt concentrations. Hence, the use of ion-exchange technologies must be evaluated on a case-by-case basis. However, further researches are being conducted on metal recovery and reuse. But, metal recovery are not economical for most industries using conventional concentrating techniques. Technologies should be evaluated that recover heavy metals from regenerating solutions of ion-exchange processes to enhance their potential economic recovery [1].

IV. FLY-ASH ZEOLITES

Fly ash, a coal combustion residue, has been used as a raw material for synthesis of micro porous aluminosiliceous minerals known as zeolites. Due to their excellent ion exchange capacity, high surface area and unique pore characteristics, zeolites have been used for removal of heavy metals such as As, Cd, Cr, Cs, Cu, Fe, Hg, Mn, Ni, Pb, Sr, W and Zn and ionic species like ammonium, chloride, fluoride, nitrate, phosphate and sulphate from industrial sludges, acid mine drainage and wastewater from domestic an industrial sources.[19]. Due to the presence of innocuous exchangeable ions, zeolites have been proven to be ideal for ion-exchange applications, such as removal of hazardous heavy metals and radioactive ions [20]. Furthermore, they are excellent sorbents, owing to their highly porous microstructure containing a large number of rigid cages and channels[21]. The heavy metal removal efficiency of fly ash zeolites is much higher as compared to raw fly ash, which can mainly be attributed to their mineralogical alteration[22]. For example, while, raw fly ash removes <8% Pb^{2+}, its zeolitized counterpart shows up to 98% removal[23].Bacterial loading on fly-ash based zeolites are used for heavy metal and phosphate removal. Landfill leachate treatment using raw fly ashes, utilization of fly ash based zeolites could yield better results. Review on studies conducted on flyash by Koshy has given insight on how initial contaminant concentration,pH and the solid to liquid ratio of the system affects the removal efficiency by precipitation and sorption units.[19].

V. SURFACTANT MODIFIED ZEOLITE

Zeolites usually have high tendency towards cation exchange. Hence, adding sorbents to zeolites builds affinity towards anions and non-polar organic compounds. They do so by building hydrophobic organic coating that promotes sorption of non-polar organic compounds.

Studies conducted by Yi Dong and his coworkers have used sorbents such as hexadecylammonium bromide (HDTMA) for modification of zeolites prepared by fly-ash. They had used the same compound for removal of Bisphenol-A(BPA) which are found in waste water treatment plants(WWTP).WWTPs are the major source of endocrine disrupting chemicals (EDC) and these surfactant modified zeolite are used in removing EDC. It had shown great adsorption capacity. This was due to pH conditions, namely alkaline pH which resulted in deprotonation of BPA to form organic anions. Low temperature and presence of NaCl improved slightly its properties.Thus we can comment that modifying zeolites using surfactants shows great adsorption capacity as compared to natural zeolites, due to its specific properties such as chemical composition, surface area and total-external cation exchange capacity. Also, it forms trilayer micelle on external surface of zeolites[24]. Although clay minerals and zeolites have permanent negative charges in their

crystal structures, which enabled them to be modified by cationic surfactants. This modification of the minerals enhanced the retention of organic pollutants [25-29].
In recent years, more research have been focused on zeolite synthesized from coal fly ash (ZFA) for water treatment, i.e for sorption of inorganic cations and anions [30-33].

SURFACTANT MODIFIED ZEOLITIES (SMZ) IN DYE INDUSTRIES:

The discharge of dye effluents to the environment, especially to water system, is becoming a major concern due to their toxicity. The origins of these dyes are usually from industries such as textiles, dyestuff manufacturing, dyeing, and printing. These dyes can consume the dissolved oxygen required by aquatic life and some of them have direct toxicity to microbial populations and even can be toxic and/or carcinogenic to mammals. A suitable surfactant modified zeolites such as Sodium dodecylbenzensulfonate (SDBS), Sodiumdodecyl sulphate(SDS), Cetylpyridinium bromide hexadecyl (CPB) and Hexadecylammonium bromide (HDTMA). was prepared by Xiaoying Jin for batch adsorption of methylene blue and Orange II. This was done so because these modified zeolites such as SDBS & SDS had better adsorption capacity for methylene blue, CPB & HDTMA for Orange II than natural zeolites. Their observations had shown that adsorbed methylene blue and Orange II decreased with increasing adsorbents while the adsorption rate increased when amount of adsorbents increased. The removal rate of Orange II on modified zeolites were much higher than that on unmodified zeolites. But natural zeolites may adsorb more methylene blue than Orange II due to its surface charges. Point to be noted is that SDBS have significant advantage because of its relative cheaper price and similar to adsorption capacity as compared to SDS. Their studies have shown potential of modified zeolites for removal of organic dyes from waste water due to their relative low price and abundance in nature.

A pilot test of SMZ as a sub-surface permeable reactive barrier for remediation of contaminated groundwater was conducted at the Large Experimental Aquifer Program (LEAP) site at the Oregon Graduate Institute (OGI) in Beaverton, OR, USA.A complete description of the pilot test is given in Bowman's article [34].Bowman had also conducted laboratory column study to evaluate the performance of SMZ in removing petroleum hydrocarbons from oilfield wastewaters. SMZ were also tested to remove pathogens from sewage-contaminated water. This was demonstrated by the works by Schulze-Makulch [35]. Based both on laboratory and field results, SMZ performed very well in removing viruses from the groundwater. The only concern is the non-selectivity of SMZ, e.g. the removal of both viruses and negatively charged ions such as bromide,which may limit the applicability of SMZ in high-salinity waters and in long-term applications.

As we have seen numerous applications of SMZ for water treatment, but main issue with SMZ still lies in its stability. It can be improved by adding surfactants other than HDTMA or it can be improved by physical methods or by calcining it.[36] More research can be done in this field and prove a great opportunity.

CONCLUSION AND FUTURE POTENTIALS

As we reviewed to how ACs and zeolites play in treating waste water, improving their properties is still garnering interest. We require efficient and effective water treatement at low cost.Activated Carbon and zeolites has their own advantages and disadvantages. So a blend of the two can combine the shortcomings of either of them. Thus to enhance the cleaning power of surfactants, we need to think about a bleand of activated carbon and zeolites for waste water treatement, irrespective of quality or nature of water. Studies are being carried over surfactant based treatements. This review can open up discussion on new type blended surfactants for water treatment.

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