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An Overview of the History of Using Adsorbents in Environment

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ABSTRACT

There are several techniques for removing contaminants from water and wastewater. In the meantime, the surface adsorption is one of the simplest, the most effective, and economical methods for wastewater treatment. This review article refers to the recent research on the removal of various contaminants with various adsorbents from water and wastewater. Knowing the conducted research and background of the research topic can greatly help the study progress and achieve new logical results. Awareness of the previous research on the subject puts the study path in the continuation of the prior studies and is nothing duplicate and separated from them. Here are some examples of research from both internal and external sources on the subject of research. The movement of groundwater due to the complexity and irregularity of boundary conditions and the heterogeneity of the porous environment can rarely be studied by analytical methods. Therefore, in order to solve groundwater problems, either numerical methods should be used or experimental models should be used in problems where the governing equations also have errors. Experimental models can be divided into two parts, physical or allegorical, which were studied in detail in previous sections. Since the equation governing the lattice models is the same as the equation governing groundwater in saturation, but its structure is not like the real porous medium, this model can therefore be considered as an allegorical model.





GRAPHICAL ABSTRACT

Introduction

Mohammad Ebrahim Olia et al. (1996) used mineral nanosorbents to separate (remove) copper metal from the effluent of textile industry [1-3]. Initially, the parameters of the adsorbent concentration, metal concentration (analyte) under analysis, pH effect, stirring speed effect, and temperature effect were investigated and optimized, and finally the desired removal was performed on the actual effluent from the fabric dyeing process. In this research, Langmuir adsorption isotherms for different states have been plotted using nanocellular mineral nanosorbents to evaluate and analyze the results [4-6]. The separation process and adsorbentanalyte interaction stage (metal sample) in 500 mL human and Jartest device was used for a sample circulation (increasing molecular dynamics of the matrix) [7]. The sample under the study to measure the extraction efficiency by the device v. Vis Two-dimensionality was measured and the parameters 1/Ce and 1/q, Ce, q were investigated. The results revealed a high efficiency of 96% in the removal of copper metal analytes in this effluent sample, in which case this

method can be applied in industry and on a larger scale [8-10].

Saleh Salehnia et al. (1995) analyzed the effect of magnetic powdered activated carbon by iron oxide nanoparticles on the removal of copper ions from an aqueous solution on a laboratory scale and in a continuous flow regime with the aim of investigating the effect of various parameters such as pH, contact time, adsorbent dose, and the initial concentrations were examined. SCM, TGA, and FT-IR spectra were utilized to investigate the properties of the synthesized adsorbent [11]. The residual copper concentration was read by a flame atomic absorption spectrum at 324.8 nm. The obtained data were described using Freundlich and Langmuir isotherm models [12]. The findings of this study indicated that the synthesized nanoparticles can remove more than 96% of copper ions at pH=10 and for 2 minutes with an absorbent dose of one gram per liter. The results of this study further revealed that the pattern of copper uptake is more consistent with the Langmuir model. According to the findings, the synthesized magnetic nanoparticles coated with activated carbon in an aqueous medium are

dispersed and can be easily separated by an external magnetic field. On the other hand, due to the presence of the activated carbon and active adsorption sites in the structure of iron oxide, the adsorption capacity has increased and these nanoparticles have indicated high efficiency in removing copper contaminants from aqueous media [13].

In 1995, Mohammad Hossein Salmani et al. studied the removal of copper ions from contaminated water by synthesized iron oxide nanoparticles in the presence of tangerine peel extract. The co-precipitation method was utilized in order to prepare iron oxide nanoparticles. Tangerine peel extract was used to prevent aggregation and reduce particle diameter, as well [14]. The effect of various parameters such as contact time, pH, initial copper ion concentration, and the adsorbent amount on the efficiency of the contaminant removal process in the discontinuous system was investigated and optimized, too. In addition, to study the adsorption process, the experimental data were analyzed by Langmuir and Freundlich adsorption models [15]. The results revealed that the percentage of copper ion removal maximized with increasing pH of the solution. Increasing the concentration of copper ions from 5 to 10 mg/l has reduced its removal percentage from 88% to 81%. The highest percentage of copper ion removal reached 92% when the amount was 0.6 g and the contaminant concentration was 5 mg/l. The results of copper adsorption experiments showed that the copper ion removal process is significantly dependent on pH and the removal percentage is inversely related to the concentration of copper ions in solution [16]. The adsorption process of this pollutant by iron oxide nanoparticles follows the Langmuir isotherm model. The results of this study demonstrated that iron oxide nanoparticles synthesized in the presence of tangerine peel extract are suitable and efficient for removing heavy metals from aqueous solutions [17-19].

The effect of different parameters on the carboxymethylcellulose efficiency of gel composite in the process of a discontinuous adsorption of copper cation was evaluated by Kowsar Sedighi Shiri et al. For this purpose, the composite was initially produced by the free radical polymerization process, and then it was used as an adsorbent [20-22]. The discontinuous adsorption experiments were divided into five categories. These experiments were performed inside glass humans. Likewise, all samples with a volume of 25 cc have been tested. The effect of various parameters such as contact time, pH, and the amount of adsorbent was investigated. In all experiments, the effect of the desired parameter was determined by changing the desired parameter and keeping the other parameters constant. The results of the experiments showed that the equilibrium of adsorption equilibrium reached its maximum value as $q_t=30.06$ mg/g in 40 minutes and the adsorption of copper cations by hydrogel in acidic environments decreased, the lowest of which was at pH=1. The absorption value q_t =3.04 mg/g has been reached. Also, with increasing gram of adsorbent, the equilibrium capacity decreased and reached $q_t=13$ mg/g. The results revealed that the adsorption of copper cation by the synthesized gel is desirable and the comparison between the correlation coefficients of different models of quasi-quadratic equation with the value of R²=0.98 was more consistent with traditional and laboratory data [23-25].



Figure 1. Investigation of the effect of contact time on the adsorption capacity of copper cations



Figure 2. A quasi-first-order synthetic model in the synthetic adsorption of copper cations

In order to remove copper ions from water by chitosan/CTAB sodium alginate granular hydrogel, Mohammad Sadegh Mansournejad et al. (1996), initially synthesized a polymer gel and the desired hydrogel was made by first pouring 60 cc of deionized water into humans and zinc. A magnet heater was placed, and then gently 3 g of sodium alginate was added to the water so that it does not clump. Mixing was continued for half an hour after all sodium alginate was poured into humans. Chitosan was added to humans after the solution was uniform [26-28]. Then, 0.5 g of CTAB surfactant was added to the mixture (alginate and chitosan). The solution was injected into calcium chloride solution. To do so, 10 g of calcium chloride was added to humans and dissolved in 1000 cc of deionized water [29]. After making calcium chloride solution, sodium alginate solution and CTAB surfactant were injected drop

by drop into the syringe and gently absorbed granules were formed. This operation was necessary to obtain the grains of the same shape and size [30]. It was further placed on the mixer for 1 hour to stabilize the adsorbent grains. After an hour, the seeds were removed from the calcium chloride solution by a strainer and after several washing steps; it was spread on the strainer with deionized water and placed at room temperature for drying. Finally, after drying, the seeds were pulverized [31]. Batch adsorption experiments and the initial concentration effect parameters were evaluated at concentrations of 50, 100, 200, 300, and 400 ppm and temperatures of 0, 25, and 50 °C. R²=0.9524 indicates that the adsorption of copper contaminant by the adsorbent is in good agreement with the Langmuir isotherm and the Langmuir isotherm separation of 0 <RL<1 indicates that the adsorption is optimal. Δ H= 13374.73, Δ S=40.816, and Δ G=1211.5638, respectively implied that the endothermic process, the absorption process system is associated with an increase in irregularity and is likely a non-spontaneous process [32].

In 1992, Farshad Givian in his study used chitosan filled with metal oxide nanoparticles to remove lead in seawater. The properties of the synthesized adsorbent were analyzed by scanning electron microscopy (SEM) and FT-IR. The effect of the adsorbent amount, contact time, the initial concentration of contaminant on adsorption amount, temperature, and pH were further investigated. The results revealed that the zinc oxide nanoparticles coated on chitosan had effectively adsorbed lead ions and the adsorption percentage was significantly impacted by the above parameters. The maximum percentage of lead uptake was 98% in the initial concentration of 3mg/l, the amount of adsorbent was 0.05 g, pH11 was 25 °C for 45 minutes, and finally the maximum percentage of cadmium was 90% in terms of the amount of adsorbent 0.05 g, pH11, was measured for 45 minutes and in a test in which led and cadmium were adsorbed in a mixture of 3 ppm. It was found that the percentage of lead uptake is 74% and the percentage of cadmium uptake is 14%. The result is that nano-coated zinc oxide on chitosan selectively absorbs lead ions, or in other words, ions [33].

In 1996, Mohammad Sadegh Mansournejad et al. studied the removal of copper ions from aqueous solutions by CTAB-modified hydrogels by adsorption in a batch system. The effect of the prominent parameters on copper removal such as pH, contact time, and initial concentration were investigated. This experiment was performed at pH 2, 3, and 6 (without any additives) and the solution without acid or base additive had the best pH and increased the adsorption capacity. In this experiment, a copper solution with the required concentrations in 100 and 250 mL Erlenmeyer was prepared and 20 mL of each solution was poured into small humans and 0.1 g of adsorbent was added to them. The experiment was performed at times 5, 15, 20, 40, and 60 minutes, all experiments were performed at laboratory temperature (25 °C). The results revealed that the equilibrium time is 20 minutes. The kinetic studies of adsorption of copper by adsorbent demonstrated that the system follows a quasi-quadratic model with R²=0.998. Studies were performed at concentrations of 50 to 400 ppm and the results indicated that with the initial concentration. increasing the adsorption efficiency primarily increased, then decreased, and the adsorption capacity further increased.



Figure 3. Effect of contact time on adsorption of copper by adsorption (adsorption capacity)

In 1996, Mohammad Sadegh Mansournejad et al. investigated the effect of surfactant, pH, and salinity of copper adsorption solution on chitosan-based polymer hydrogel by adsorption in a batch system. The CTAB-modified polymer hydrogels were utilized as adsorbents. Five-

aqueous copper sulfate salt (CuSo₄.5H₂O) from Merck Company was used to prepare copper (II) solution. One molar HCl solution was used to adjust the pH. The copper solution with a concentration of 200 ppm was prepared in 250 mL erlens and 20 mL of each solution was poured into small humans and 0.1 g of adsorbent was added to them. Parameters affecting copper removal such as the effect of surfactant, pH, and salinity of the solution were investigated. This experiment was performed in different amounts of CTAB surfactant 0.005, 0.01, and 0.75 and without adding this substance at laboratory temperature. After 20 minutes, the samples were filtered using filter paper and 25% by weight ammonia reagent was added to them and the concentration of solutions. The absorbance of the samples was measured using а spectrophotometer. Concentration of 0.005 with adsorption percentage Re=99.976 increased and improved adsorption. The experiment was performed at pH 2, 3, and 6 (without any additives) where the solution without acid or base enhancer has the best pH and increases the adsorption capacity. The salinity effect test of CTAB, CaCl₂, BaCl₂, and NaCl ions, each of which was added to the solution at the rate of 0.01 g, as well as a non-additive solution was investigated that the percentage of adsorption without any additives improved and increased the adsorption [34].

In 1996, a new hydrogel was made by Kowsar Sedighi Shiri et al. using carboxymethylcellulose with different amounts of bentonite (0.1 and 0.5) g through free radical polymerization method and it was evaluated as adsorbent of copper cation from an aqueous solution. The synthesis of the hydrogel composite as well as the effect of various parameters such as pH contact time and adsorbent amount were investigated discontinuously. The results revealed that the adsorption of copper cations by hydrogel was reduced in two samples compared to the acidic environments, the lowest of which at pH=1

reached the adsorption values of q_t =16.31 and 14.36 mg/l. Likewise, the equilibrium adsorption capacity decreased with increasing adsorbent in the two samples compared, the minimum value of which is q_t =12 g/g. While the removal percentage has increased, the maximum value is q_t = 57.7. The adsorption of copper cation by the synthesized gel is favorable and the comparison between the correlation coefficients of different models demonstrated that in both samples the quasi-quadratic equation with values of R²=0.99 and R²=0.97 were more consistent with the traditional and laboratory data, respectively.

A review of past studies of fixed bed columns

To perform a continuous process in the field of adsorption, one of the most widely used equipment used in this field is fixed bed columns. Much research has been carried out on the use of fixed bed columns.

In 2014, Ai Phing Lim et al. conducted their study on a continuous bed column and modeled adsorption, removal of cadmium II and lead II ions in an aqueous solution by a dead limestone skeleton. In the Thomas and Adam-Bohart and Eun Nelson column absorption model, the Thomas model was more consistent with the cadmium correlated curve data. The Ion Nelson model was chosen to estimate 50% of the progress time by the fixed bed column, and the progression time estimate is provided for columns that were not perfectly used during operation. The Adam-Bohart model can be used for the initial part of adsorption with equilibrium concentration data. The saturation index of aragonite and calcite is depicted in the figure when calcium dissolution occurred in aqueous solution. The experimental data and theory were significantly related to each other, which demonstrated that the experimental data were well matched with the modeling data. Both experimental and theoretical data trends were reasonably dependent on each other due to the column parameters and CS materials, which indicates a significant trend of experimental data and modeling process. The effective column parameters in this work were presented in the form of a table, and then the diagrams comparing the cadmium removal with the traditional models and indicating the trend of both experimental and theoretical data in relation to the column parameters and CS components were further given.

In 2017, Mohammad Kavand et al. studied an improved dynamic diffusion model in a fixedcolumn adsorption film on heavy metal ions, single-component, and multi-component systems. In this paper, the adsorption of heavy metal ions such as cadmium, lead, and nickel on commercially activated carbon in single and multi-component stable columns is investigated. The effects of several parameters such as initial concentration, flow velocity, and bed height, as well as the traditional explanation of adsorption and evaluation of mass transfer coefficients, the precise calculation methods were performed to determine the contribution of each mass transfer resistance. The model has been successful for mass transfer parameters in multi-component and single-component systems under different operating conditions. The number of biobs increases with maximizing the current and initial concentration, which indicates an increase in resistance to penetration into the particle.

In 2010, Ensar Oguz et al. investigated the adsorption potential of sunflower husk to remove Cu2 + from aqueous solution using a fixed bed adsorption column. The effects of the initial concentration of $Cu^{2+}(20-60 \text{ mg/g})$, feed flow rate (9-21 mL/min), bed height (5-15 cm), input pH (3-5/6), and particle size (2-2 mm), (1-5-0), (0.5-0.5) were investigated on the progress of the characteristics of the adsorption system. The highest laboratory bed capacity and theory were 25.95 and 26.22 mg/g in the initial concentration of Cu^{2+} , 60 mg/l was the highest bed at 5 cm and the flow rate was 5 mL/min and pH were 5.6 and the particle size was 0.5-25 mm. The

relationships were obtained between calculated and observed data. According to the ANN model, the coefficient of determination is 0.986 and the mean of the second root error is 0.018. The results revealed that sunflower residue is a suitable adsorbent for removing Cu²⁺ copper ion from aqueous solution.

Using the adsorption column, Kowsar Sedighi Shiri et al. (1996) evaluated the effect of different parameters on the performance of the carboxymethylcellulose cell composite of poly bentonite in the continuous adsorption process of copper cation. For this purpose, the composite was primarily produced by the free radical polymerization process, and then used as an adsorbent. Adsorption experiments were performed using a glass column 25 cm long and 1.5 cm in diameter at ambient temperature. The adsorption capacity of the column to remove copper under different operating conditions was investigated. The values of 3, 6, and 9 cm were selected to test the effect of bed height and mL, 4.5, 7.5, and 16 mL/min were selected to test the effect of input flow and 49.5, 270, and 454 mg/L were selected to test the effect of initial concentration of copper solution. The copper solution was transferred from the top into the column. Samples were taken at regular intervals from the column output. The amount of copper in the samples was analyzed by spectrophotometer. Failure curves were further plotted for the experiments performed. The effect of different parameters of the initial ion concentration, adsorbent column height, injection flow on adsorption efficiency, wear time, and MTZ was investigated. The failure curves were plotted to evaluate the effect of each parameter. The results showed that with increasing the height at the maximum height of 9 cm and at the lowest flow, the wear time increased. Besides, with increasing flow, the mass transfer area decreased by 5.2858 cm and the refractive curve is sharp at high input concentrations. This indicates that the mass transfer area is small.

In the kinetic study of adsorption of copper pollutants by composites (chitosan, sodium alginate, and CTAB) on a fixed bed of adsorption experiments using a glass column 25 cm long and 1.5 cm in diameter at 25 °C in 1996 by Mohammad Sadegh Mansournejad et al., to find out the effect of initial concentration of input solution on the adsorption capacity of the column, values of concentrations of 10, 30, and 50 ppm for input solution to the column at a height of 3 cm and a flow rate of 12 mL/min were selected at regular intervals. The output solution was sampled from the column and 25% by weight ammonia reagent was added to them and the concentration of the solutions was measured using a spectrophotometer. The fracture curves were plotted to investigate the effect of concentration parameter and synthetic models. The results of the initial concentration effect test revealed that by increasing the concentration, the parameters (qe, qt, and Wt) increased, respectively, which showed that with increasing the adsorbed concentration, the load of contaminants with the substrate increased and the driving force of mass transfer increased. Comparing the values of correlation coefficients of the kinetic models applied to the laboratory data demonstrated that among the three traditional models studied, the Thomas model and the Eun-Nelson model with a correlation coefficient of R²=0.95 were more consistent with the laboratory data.

Kowsar Sedighi Shiri *et al.* (1996) used acrylamide in the study of gel nanocomposites based on carboxymethylcellulose, AMS, and bentonite nanoparticles using free radical polymerization method and used an aqueous solution as adsorbent of copper cation, as well. The morphology of the adsorbent was evaluated by SCM analysis. Then, considering that different parameters can affect the continuous adsorption process, the effect of different parameters of the initial ion concentration, adsorbent column height, and injection flow on adsorption

efficiency was investigated. During the experiment, the concentration of copper ions in solution was measured aqueous by а spectrophotometer. According to the laboratory data, a kinetic study was performed on the data and Adams-Bohart, Thomas and Eun-Nelson kinetic models were applied to predict the failure curves. The values of the correlation coefficients illustrated that the Thomas Vion-Nelson models with the highest values of $R^2=0.93$ and $R^2=0.92$, respectively, are in good agreement with the laboratory data.

LiU et al. investigated the removal of lead ions in a continuous process in a fixed bed column by a group of amines and powdered carboxyl as an adsorbent from ethanol gassing solution. The results revealed that increasing the volume of flow from 0.12 to 0.34 mL increased the amount of lead absorption from 54.9% to 2.3%. However, increasing the mass flow to 3.6 mL/min did not significantly change the lead removal percentage, indicating that the solution was still turbulent. The temperature results also showed that at a temperature of 25 °C to 65 °C in a solution of gassing ethanol with an initial value of 1.7 mg/kg to 0.26-0.05 mg/kg after a period of adsorption that the amount of 8 84.8% to 96.8%, which indicates that increasing the temperature could enhance the adsorption of lead by the adsorbent. B.Albadari et al. examined the technical study of chromium adsorption for the production of activated carbon in lignin. In this study, continuous removal of chromium (VI) ion by activated hydroxide in lignin by Microc dumm method was used to determine the adsorption parameters by the adsorbent. Studies have implied that the adsorption capacity of this adsorbent is strongly dependent on pH and the adsorption of chromium decreases with increasing pH, which was similar to the trend when increasing the flow rate and ionic strength. The experimental data were analyzed with the Thomas-Fermi model of Dose-Response model for variable initial pH, ionic strength, initial

concentration of chromium ion (VI), and adsorbent mass (PAL). The modified reaction dose change model is able to express chromium uptake in the process (PAL).

Concluding

Using a homogeneous precipitation method, the synthesis by homogeneous loading methods was investigated by BET (Brunauer-Emmett-Teller), XRD (X-ray diffraction), and XPS (electronic Uscan) methods. The adsorption of the elemental measured aluminum mercury was bv nanoparticles under nitrogen and simulated gas space on a fixed-scale fixed device. The effect of different gases on the removal performance of Hg⁺ was analyzed using nano-ZnO. The results reveal that the efficiency of nano-ZnO mercury in nitrogen space is relatively low. The presence of H²S significantly removes Hg0 by nano-ZnO, and mercury removal efficiency can be the maintained for a long time even after the passage of H²S has stopped. The presence of CO and H² removes Hg^+ by removing nano-ZnO sulfurization. As the temperature increases, the formation of elemental sulfur at the nanoparticle surface gradually decreases, leading to the removal of Hg⁺. The present work reduces an indirect and benign environment for the synthesis of ZnO nanomaterials using sugar candy. The synthesized nanomaterials were characterized by XRD, SEM, TEM, BET, EDS, and FTIR. The UV-visible and photoluminescence studies were further performed to identify the adsorption properties of the synthesized nanomaterials. It is clear from adsorption studies that ZnO synthesis should be applied as an effective adsorbent to remove Pb (II) and Cd (II). Kinetic data follow a quasi-second-order model. Equilibrium in 120 minutes and isotherms are Sips>Langmuir>Freundlich, respectively. Adsorption studies of desorption over 6 cycles indicate the ability and frequent use of adsorbents to remove Pb (ll) and Cd (ll) from aqueous solutions. The practical efficiency and usefulness of the adsorbent were also tested using real industrial wastewater. The results of cytotoxicity indicate that ZnO is compatible at lower concentrations of the environment and is used as an ecological organic nanoparticle for industrial and environmental applications.

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