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Investigation of Adsorption Capacity of Metal-Organic Polymers

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Abstract—In this work, we studied a biocompatible hybrid material based on iron (III) and muconic acid oxoclusters. It has been shown that coordination polymers are a promising class as functional materials for various purposes (as sorbents, catalysts, conductors, storage materials, etc.).

The adsorption capacity of the obtained adsorbent for removing dyes from the prepared solution was in the following order: CR> MB> MV. From the results of the study, we can conclude that the dye Congo red is best suited for adsorption by the coordination polymer.

The maximum absorption of the dye on organometallic coordination polymers occurs in the pH range 5 - 7 with adsorption of ~ 90%, which is important for the potential practical application of such coordination polymers as carriers for drug delivery.

Keywords—Polymers, adsorption, adsorption capacity, drug delivery, dyes, stability.

I. Introduction

Most of the existing drug carrier materials exhibit poor drug capacity (usually less than 5 wt%) or rapid release of a fraction of the drug [1]-[14]. In this aspect, it was interesting to consider the possibility of using the resulting system as a carrier for targeted drug delivery.

Among the wide range of investigated nanoscale materials for various environmental and biomedical applications, magnetic nanoparticles have received considerable attention due to their intrinsic magnetic properties, making them successful as magnetically reduced catalysts, drug delivery agents, anti-cancer materials, magnetic resonance imaging, etc. This class of nanomaterials includes metallic, bimetallic nanoparticles, metal oxides, ferrites, and superparamagnetic iron oxide nanoparticles. Magnetic nanoparticles and nanocomposites have generated significant scientific and technological interest due to their potential applications in biomedicine, information technology, magnetic resonance imaging, catalysis, telecommunications, and environmental restoration [1]-[14]. Magnetic nanocomposites usually contain magnetic nanoparticles embedded in a non-magnetic or magnetic matrix. However, magnetic nanoparticles dispersed in composites usually have a strong tendency to form agglomerates to reduce the energy associated with the high surface area to volume ratio of nanosized particles [15-14]. To avoid aggregation of magnetic nanoparticles, protection strategies have been developed to chemically stabilize unprotected magnetic nanoparticles by grafting or coating with organic species, including surfactants or polymers, or coating with an inorganic layer such as silicon dioxide or carbon. Combining these functionalized magnetic nanoparticles in a polymer or other matrices to develop magnetic nanocomposite materials has proven to be more efficient [25]-[32]. There are mainly four types of magnetic nanocomposites, i.e. inorganic core nanocomposites, self-assembled nanocomposites, silica-based magnetic nanocomposites, and organic-inorganic nanocomposites [33]-[50]. Among them, organic inorganic magnetic nanocomposites have become more interesting due to the combination of the unique properties of the organic and inorganic components in one material. Hybrid organic inorganic magnetic nanocomposite materials can be obtained in situ, ex situ, microwave exposure, coprecipitation, melt mixing, ceramic glass treatment, and plasma polymerization methods.

The properties of the interface or interfacial zone, first of all, the adhesive interaction between the fiber and the matrix,

determine the level of properties of composites and their retention during operation. Local stresses in the composite reach their maximum values just near or directly at the interface, where material destruction usually begins. The interface must have certain properties to ensure efficient transfer of the mechanical load from the matrix to the fiber. The adhesion bond at the interface should not be destroyed under the action of thermal and shrinkage stresses arising from the difference in the temperature coefficients of linear expansion of the matrix and fiber or as a result of chemical shrinkage of the binder during its curing.

Magnetic nanoparticles can act as a new class of non-toxic and effective flame retardants. Fe_3O_4 enhanced both thermal stability and flame retardant properties of polyvinyl alcohol. Nanoparticles were synthesized by a simple precipitation reaction without using an inert atmosphere at room temperature. The nanoparticles exhibited ferromagnetic behavior at room temperature. To obtain a magnetic nanocomposite, Fe_3O_4 nanoparticles were added to polyvinyl alcohol. Dispersed nanoparticles play the role of a magnetic barrier layer, which slows down the volatilization of the product and prevents the penetration of oxygen into the sample during polymer decomposition.

The effectiveness of magnetic nanoparticles for specific biomedical applications depends on the magnetic properties of the particles, their size, and most importantly, the surface chemistry [1]-[9]. Of particular importance in the biomedical application of colloidal magnetic systems is their stability, which depends on the effective stabilization of nanoparticles, which would ensure the presence of individual nanoparticles rather than agglomerates in solutions, while not significantly affecting the magnetic properties of the material and preserving the particle surface suitable for further functionalization [10]-[18].

For this purpose, magnetite nanoparticles of the "core-shell" type are widely used, having an inner core of iron oxides (Fe_3O_4) with an outer protective shell of silicon dioxide (SiO_2). SiO_2-based coatings solve a double problem: first, they prevent the aggregation of nanoparticles and the oxidation of magnetite (both problems are fundamental in the transport of nanomaterials through the bloodstream); second, they allow the surface to be modified with various specific ligands for biomedical applications [51]-[74].

In order to make sure that the obtained coordination polymer based on oxo-centered iron complexes possesses the required storage capacities, their adsorption capacity was studied. For this, organic dyes methylene blue (MB), Congo red (CR), and methyl violet (MV) were used as model systems. The choice of these types of dyes is due, on the one hand, to the different nature of their existence in solution: Congo red in solution is in the anionic form, methylene blue and methyl violet are in the cationic form, on the other hand, by a different type of structure, which will make it possible in the future to select medicinal drugs close to them in properties.

II. Study of the Sorption Activity of Coordination Polymers

Modern composites have not only a wide range of physical and mechanical properties, but are also capable of directionally changing them, for example, increasing fracture toughness, regulating rigidity, strength, and other properties. These possibilities are expanded when fibers of different nature and geometry are used in composites, i.e., when creating hybrid composites. In addition, these materials are characterized by the appearance of a synergistic effect (coordinated joint action of several factors in one direction).

In order to derive the equation of the adsorption isotherm, a number of simplifications are introduced. All the places where the adsorbed particles are fixed are the same, and adsorption on one of them does not affect the state of the other. The interaction between the adsorbed particles is negligible. The adsorption layer is monomolecular, i.e. it consists of one layer of molecules. In this case, the bond of the adsorbate with the adsorbent is sufficiently strong, which excludes the movement of the adsorption complex along the surface of the adsorbent (localized adsorption).

Active (well absorbing) adsorbents have a very large specific surface area. For example, the absorbers most often used for scientific purposes and industrial practice - activated carbon, silica gel, zeolites – have s_0 up to several hundred and even thousands of square meters per 1 g.

The ability of the adsorbent to absorb the adsorbate is characterized by the amount of adsorption. The amount of adsorption is the excess mass of the adsorbate in the boundary layer over its mass in an equal volume of the environment, referred to the unit surface of the adsorbent.

Sometimes the adsorption value is expressed in moles of adsorbate per 1 m^2 (or 1 cm^2) of the adsorbent surface. Since quite often the surface of the adsorbent is unknown, the value of adsorption is expressed in moles of adsorbate per 1 g of adsorbent (mol/g). It is customary to evaluate the process of toxin sorption by the adsorbing surface using the curves of Langmuir sorption isotherms.

Silanol binding agents are applied directly to the surface of Fe_3O_4 nanoparticles by copolymerization of monomers or by direct silanization. The developed surface of nanoparticles leads to a high density of surface functional groups [48]-[57], which can fix a large number of biologically active substances [32]. The most common way to obtain LF Fe_3O_4/SiO_2 with a core-shell structure is the sol-gel method (Stober method), which consists in hydrolysis and polycondensation under alkaline conditions in ethanol [33].

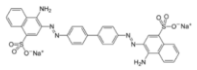
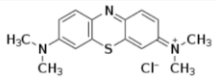
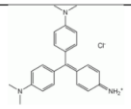
Coatings on nano and micro-sized particles can serve for many purposes. First of all, modification of the surface with coatings makes it possible to make the particles compatible with various matrixes [14]-[30]. For medical purposes, the biocompatibility with the environments of a living organism is of crucial importance. It is equally important that coatings can significantly enhance or decrease the sorption properties of magnetically controlled sorbents. This provides prerequisites

for the creation of magnetically controlled particles with specific sorption properties. It is also known that the coatings prevent the core from leaching out. The presence of a coating also often facilitates the stabilization of particles in an environment with an alkaline pH or significant salt concentration. For example, the isoelectric point of SiO_2 is reached at pH 2-3. Therefore, the particles coated with silica are negatively charged at the pH of the blood, which causes electrostatic repulsion, which avoids the formation of clumps.

III. Results and Discussion

Organic dyes were used for the study: methylene blue (MB), Congo red (CR) and methyl violet (MV). Their structural formulas and wavelengths are presented in Table 1.

Table 1. Characteristics of the studied dyes.

Dye	Formula
CR λ_{max} = 492 nm	
MB λ_{max} = 664 nm	
MV λ_{max} = 584 nm	

The quantitative values of adsorption for different organic dyes differ significantly, the results obtained are shown in Table 2.

Table 2. Quantitative values of adsorption at different temperatures.

Dye	Adsorption values at different temperatures, mg/g	
	283 K	308 K
CR	39.6	39.4
MB	37.6	36.4
MV	36.6	36.2

The adsorption efficiency reaches almost 98% for Congo red, 96.2% for methylene blue and 92% for methyl violet, which leads to almost complete discoloration of dye solutions (Figure 1).

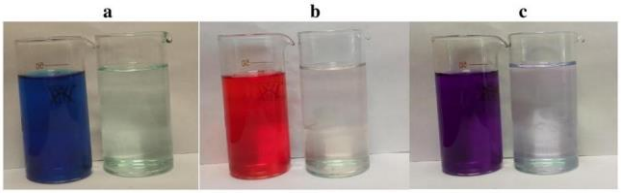


Fig. 1 Color of the dye solution methylene blue (a), Congo red (b) and methyl violet (c) before and after adsorption.

The obtained adsorption characteristics in relation to dyes at different adsorption temperatures are presented in the form of dependences in Figures 2 and 3. Figure 2 shows the values (qt) of the dye (CR, MB, MV), adsorbed on the adsorbent, depending on the adsorption time t. It can be seen that the adsorption capacity of the obtained compound rapidly increases in the initial period of contact time, and then becomes slower. In this case, rapid diffusion to the outer surface is accompanied by rapid diffusion into the pores of the matrix, which leads to the rapid achievement of equilibrium [15]-[26]. The adsorption capacity of the obtained adsorbent for removing dyes from the prepared solution was in the following order: CR> MB> MV, as shown in Figure 3.

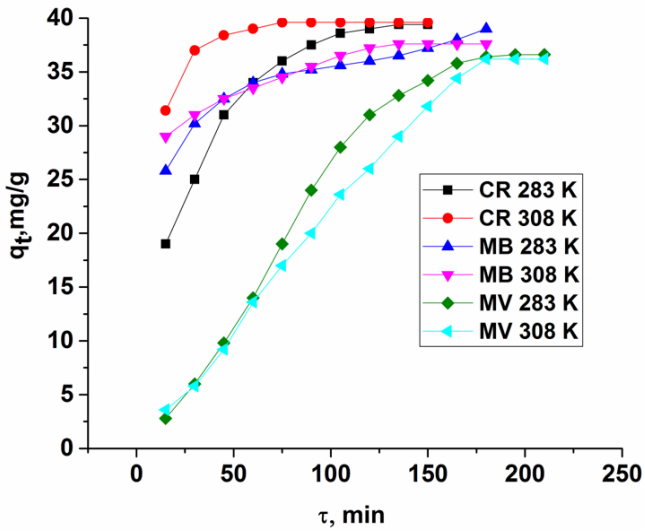


Fig. 2 Relationship between adsorbed amounts (qt) of dyes and time.

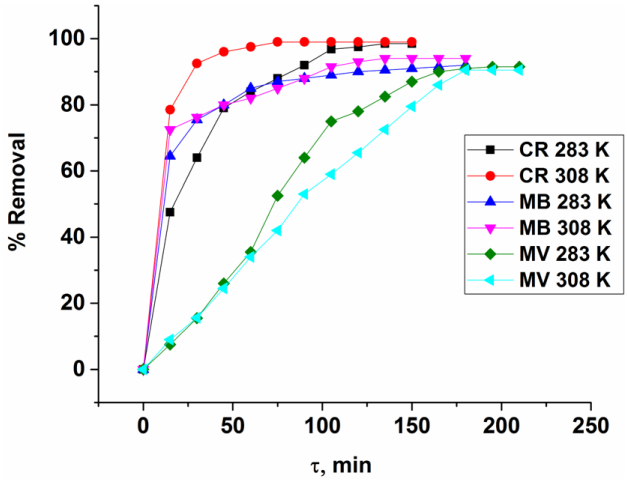


Fig. 3 Influence of contact time on the adsorption of organic dyes.

From the results of the study, presented in the Figures 2 and 3, we can conclude that the dye Congo red is best suited for adsorption by the coordination polymer.

For a nanoscale drug delivery device to be effective, premature release of the therapeutic drug must be minimized by allowing the drug to travel to the site of disease within the body to a specific target [27]-[34]. This release of the drug attached to surfaces by coordination or physical adsorption can occur at different pH values. From this point of view, it is

necessary to check in what pH range the release of the adsorbed substance can occur. The results obtained for the study of the effect of pH are shown in Figures 4 and 5.

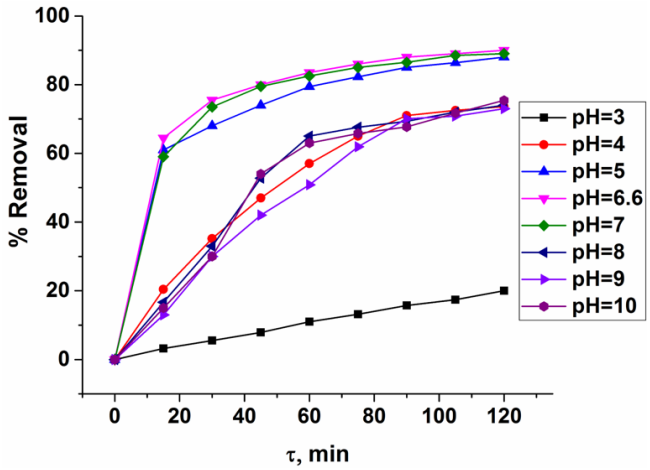


Fig. 4 Dependence of the degree of adsorption of methylene blue on pH.

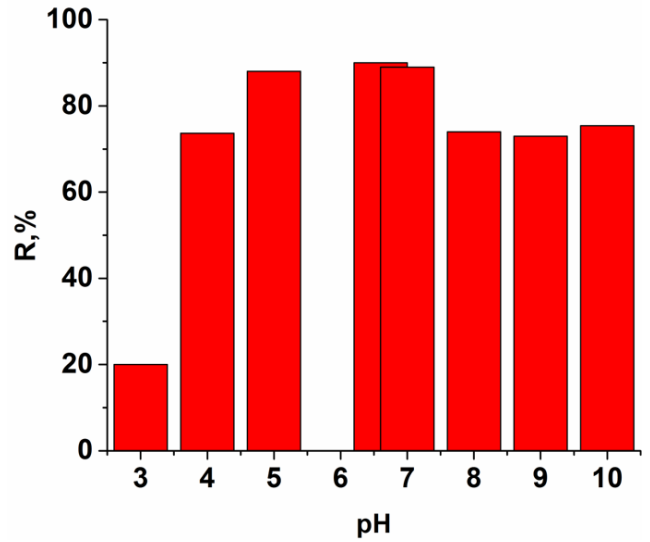


Fig. 5 Maximum degree of adsorption of methylene blue on iron muconate, depending on pH.

From the Figures 4 and 5 it is seen, that the maximum absorption of the dye on organometallic coordination polymers occurs in the pH range 5 - 7 with adsorption of ~ 90%, which is important for the potential use of such coordination polymers as carriers for drug delivery [35-40].

IV. Conclusion

The resulting mesoporous adsorbent shows a good ability to adsorb organic dyes such as methylene blue (MB), congo red (CR), and methyl violet (MV), which makes it possible to direct further studies to consider the possibility of using the obtained systems for targeted drug delivery.

It can be seen that the adsorption capacity of the obtained compound rapidly increases in the initial period of contact time, and then becomes slower. In this case, rapid diffusion to the outer surface is accompanied by rapid diffusion into the pores of the matrix, which leads to the rapid achievement of equilibrium. The adsorption capacity of the obtained adsorbent for removing dyes from the prepared solution was in the following order: CR> MB> MV. From the results of the study, we can conclude that the dye Congo red is best suited for adsorption by the coordination polymer.

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