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Review Paper

PURIFICATION OF CONTAMINATED WASTEWATER WITH THE HELP OF GRAPHENE COMPOSITES WITH HYDROGELS

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Abstract. *Lack of clean water requires the use of new wastewater treatment technologies. Adsorption is a simple and effective method for removing contaminants from contaminated water. Graphene composites with hydrogels have found application in wastewater treatment because they have unique properties such as porous structure, unique morphology, good rheological properties, non-toxicity, etc. The paper presents a literature overview of potential solutions to wastewater treatment using composite graphene and graphene oxide with hydrogel-like adsorbents. The mentioned composite compounds have been used in the treatment or elimination of various hazardous substances. In this work, we have investigated the possible adsorption of different classes of colored pollutants (paints) and pesticides (both organic and inorganic).*

Key words: *contaminated water, pesticides, dyes, composites graphene-hydrogels*

1. INTRODUCTION

Water, as a source of life, is of greatest importance for living beings. However, the fresh water that people use every day occupies only about 0.007% of all water on Earth, which triggers the issue of the lack of clean water. The current problem is the shortage of drinking water, which according to the World Bank report, 2.0 billion people face. The mentioned shortage could reach 4.6 billion by 2080 if the trend of negligence towards water handling continues [1]. The generation of industrial, hazardous and chemical waste and its improper disposal further encourage the lack of safe and clean drinking water. In most developing countries, according to the UN Water Organization, polluted water is discharged into rivers, lakes, and seas. Almost 90% of contaminated water ends up in the recipient, without any prior treatment [2, 3]. In this paper, the influence of paints, pesticides and heavy metals on water quality as well as the adsorption of pollutants from

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water through the composite graphene hydrogel have been presented. Composites generally consist of two or more materials that differ in properties [4]. Assembled, they usually make one material with a set of properties, but also with new characteristics [5]. Graphene has been shown to significantly improve the electrical, mechanical and thermal properties of traditional polymer hydrogels, making the resulting composite an excellent adsorbent. Graphene composites with hydrogel have found great application in the adsorption of pollutants from wastewater [6].

2. PESTICIDES

Modern agricultural production cannot be imagined without chemical plant protection. The use of pesticides leads to contamination of both soil and water. A lot of effort and work has been put into minimizing water pollution. The green method of water purification, efficient technology, reduced energy use and environmental protection are still being sought. There are many biological, chemical and physical methods of water purification, while adsorption has been suggested as one of the best. Both adsorption of pesticides and controlled release are processes that need great attention since they determine the fate of pesticides in water. Among many materials which function as pesticide adsorbents are composite materials.

Pesticides are a group of agrochemicals that, in addition to their benefits, exhibit many side effects. When assessing the harmful effects, the central place belongs to the danger of environmental pollution, primarily soil, leachate, as well as the recipients themselves, who are reached through leachate [7, 8]. Therefore, all tests in this area aim, above all, to obtain the data that might allow hazards assessment, i.e. risk of pesticide application.

By definition, a pesticide is a compound or mixture of compounds intended to prevent, destroy, repel or mitigate pests [9]. Pesticides can be classified according to the organism they act on (target organism), chemical structure and physical condition. The most common division of pesticides is according to their purpose, i.e. the organism on which they have impact on:

- insecticides- agents for controlling harmful insects,
- fungicides,
- bactericides - agents to control bacteria,
- herbicides- weed control agents,
- plant growth regulators [10].

Extensive use of pesticides to increase yields in agriculture and intensive development of new chemicals has drastically increased the number and quantities of agrochemicals present in the environment. The harmful effects of pesticides can be observed in both terrestrial and aquatic systems that affect both flora and fauna [11]. As a consequence of the chemical residues of pesticides, human health is also endangered. Negative effects also occur in many non-target organisms, which are benign to the environment [12]. With the advent of newer technologies and advances in materials science, smart materials are being developed that contribute to the preservation of ecosystems, because materials can be manipulated and adapted to certain desired actions.

2.1. Challenges in pesticide management

Agriculture is among the most important sectors, since it produces and provides raw materials for the food and livestock industry. The scarcity of natural resources and population growth in the world requires sustainable, environmentally friendly, and efficient economic and agricultural development. It is necessary to protect agricultural goods by using pesticides. Approximately 90% of pesticides end up in the ecosystem. This unwanted loss of pesticides leads to many diversities in nature, resistance to pests and diseases, changes soil chemistry with impaired nitrogen fixation, impairs soil biodiversity and increases the rate of bioaccumulation [13]. Moreover, excessive use of pesticides has an impact on the entire population, especially on children who can suffer from skin irritation and immune system deficiency. The EPA study from 2012 showed that the most commonly used conventional pesticides such as Glyphosate, Atrazine and Metolachlor-S might be associated with health problems such as cancer, abortion and human hormone disorders [14]. Some pesticides are characterized by being excessively persistent in the environment and belong to the class of persistent organic pollutants (POPs), including organochlorine and organophosphates (OC and OP). Several studies indicate the presence of organophosphate and organochlorine compounds in aqueous systems [15], which are usually contaminated several weeks after insecticide application. The prevalence of organophosphate insecticides in the environment and the presence in spring waters pose a serious threat to wildlife, as well as human health [16]. Organophosphates in the environment are subject to natural degradation, which is mostly through homogeneous and heterogeneous hydrolysis [17]. Insecticides, namely, Aldrin, Endrin, Mirek and Tokaphene are resistant to degradation due to their lipophilic nature. They can accumulate in the adipose tissue of organisms and biomagnify along the food chain [18].

Challenges in developing new pest control strategies include not only identifying new active compounds, but also improving the delivery of pesticides at the biological level. The development of a new strategy for the delivery of active components is important [19], therefore it is important to develop “smart systems” for dosed release or adsorption of pesticides in order to reduce the negative impact. The pesticide dosing system resembles the dosing of drugs from a carrier [20]. A meta-analysis comprising 155 scientific papers, came to the conclusion that the most investigations were conducted in Asia, 28% in China and 20% in India, out of the total number of papers analyzed, followed by the United States (20% as well). Half of the analyzed papers were related to controlled release of insecticides, approximately 30% of the papers were related to fungicides, while herbicides were analyzed in 15% of papers [21]. Since pesticides end their cycle mainly in soil or water, deterioration of water quality and the unavailability of drinking water represent key challenges worldwide. Removal of toxic organic and inorganic pollutants from water is a necessity for a clean environment in response to water scarcity, as well as for human society. Adsorption-based water technologies are among the most desirable and are most commonly used, due to their high efficiency and low cost [22]. However, the materials used for the adsorption of pesticides, as well as for their degradation, must meet several conditions. Above all, they should be porous, have a high absorption power, and be degradable.

2.2. Hydrogels and graphene hydrogel composites as adsorbents and/or in controlled pesticide release

Polymer formulations used for agricultural applications are easier than those used for drug delivery, which makes the final product commercially viable [23]. One of the important preconditions for the synthesis of polymers that would be used for controlled release or for adsorption of pesticides is that as little pesticide as possible should remain in the environment [24]. In the polymer network itself, they are present in the form of microcapsules or granules. Polymer systems that can be used as carriers of active substances or as adsorbents can be broadly divided into two categories, depending on the interaction of active substances or biologically active substances with polymeric materials, in order to obtain the desired release profile. These interactions can be either a physical combination, in which the polymer matrix acts as a rate-controlling medium or a chemical interaction, in which the polymer acts as a carrier for the active substance. However, many factors and considerations dictate the choice of monomer as a potential polymeric adsorbent or carrier to be used in a given scenario in order to achieve the desired effect while at the same time leading to minimal biological or environmental side effects. These include the route of action and the chemical nature of the active substance, their physicochemical interactions with the matrix, the nature of the polymer matrix (thermoplastic or thermosensitive), thermal behavior, lightness and stability of the formulation, desired shape and size of the final product, price, desired release kinetics, method of application, etc. [25]. One of the possible techniques in the controlled-release of pesticides is the encapsulation of pesticides in a polymer matrix, which allows prolonged pesticide activity [23]. In addition, the polymer matrix can later play a role as compost after decomposition [26]. Various synthetic, semi-synthetic and polymers of natural origin are often used to form such formulations. The characteristics of synthetic polymers are often improved to be environmentally friendly, biodegradable and have better mechanical properties [24]. Two types of polymer-based nanoformulations can be distinguished - polymer nanospheres and nanocapsules. For the first one, the distribution is not specified, while the second one has a core-shell structure that can act as a reservoir for a pesticide dissolved in a polar or non-polar solvent [27]. Importantly, nanocapsules have advantages over larger capsules (e.g., spray solution stability, increased intake, increased spray area, and reduced phytotoxicity due to more homogeneous distribution). Carbon-based nanomaterials are gaining popularity as nano adsorbent for water purification due to properties that depend on size and shape, environmentally friendly nature, abundance and ease of handling. In recent years, carbon nanomaterials (CNM), such as graphene and derivatives, carbon nanotubes, carbon nanofibers, nanoporous carbon, fullerenes, graphite carbon nitride and nanodiamonds, have been used extensively as adsorbents due to their outstanding surface properties, simple modification, large modifications, chemical stability, porosity, low density, ease of regeneration and reuse. Graphene oxides and other oxidized carbons show excellent adsorption of cationic and basic compounds through electrostatic and hydrogen-related interactions. Moreover, graphene oxide has hydrophobic surfaces and offers high adsorption through strong π-π interactions.

Many reviews have analyzed the adsorption potential of carbon and polymer-based materials [27]. For adsorption to be effective, the adsorbent must have a large surface area, many active surfaces, excellent mechanical stability, and be economically viable and environmentally friendly [29]. The agricultural sector, like many other sectors, is experiencing incredible growth in the use of nanotechnology. Just in the period from 2000 to 2013, over 3000 patents were made, 60 review papers and 25 reports and reviews were published [30] aiming to reduce unnecessary retention of pesticides in the soil, in order to avoid nutrient loss, but also to sustainably manage better yields [31]. Green and Beestman [32] in their paper gave a good overview of polymer patents used in the controlled release of pesticides. Manufacturers of polymer and pesticide formulations tend to ensure that the best additives are used on their products in order to have the best possible characteristics of the final product. The goal is to make handling and application easier but to improve performance. Glyphosate is the most commonly used pesticide in such formulations, patents with glyphosate are issued monthly, and manufacturers introduce new formulations annually. Today, the practice is to control the pH value that would lead to the desired and adequate release of pesticides [33]. However, sometimes an increased pH value is needed to release pesticides, sometimes lowered, and sometimes buffered to a neutral value. The pH value is defined by the monomers, ie the synthesized polymer used as a carrier and the pesticide to be released from the formulation. Glyphosate is currently sold in the form of an acid with a pH of less than 2, mono-salts with a pH of 4.4 to 5 and as a formulation of two salts with a neutral pH close to 6.7 [34]. Recently, a pesticide model by solvent evaporation using various polymers such as ethylcellulose, cellulose acetate butyrate butyryl and poly(methyl methacrylate) to prepare microspheres filled with pesticides was designed. The microparticles were then characterized by scanning electron microscopy and infrared spectroscopy, determining the size and particle size distribution. Pesticide release was determined in water at 25 °C. The effects of the nature of the matrix, double encapsulation and some of the process parameters such as mixing speed, the relationship between polymer and solvent, but also the relationship between pesticides and polymers were also analysed [35]. Chin et al. developed a poly(methyl methacrylate)-poly(ethylene glycol)-poly(vinyl pyrrolidone) suspension to encapsulate carbofuran. They then conducted a study focusing on the efficacy and stability of the diamond moth with two types of carbofuran formulations; microsuspension (commercial) and nanosuspension. They found that carbofuran had similar efficacy at a lower dose for the synthesized nanosystem compared to the micro suspension [36]. Shakil et al. [37] have proposed a method of preparation using poly(ethylene glycol) and various copolymers for the controlled release of insecticides. Amphiphilic polymers based on poly(ethylene glycol) were enzymatically synthesized with dimethyl 5-hydroxyisophthalate as a crosslinker. Polymer characterization was performed using 1H and 13C-NMR spectra. Desorption of carbofuran from different formulations varied from 94.2 to 99.5%. The release of carbofuran in this study followed first-order kinetics while the half-life values (t1/2- time required to release 50% of carbofuran from the carrier) of carbofuran of different controlled release formulations ranged from 7.5 to 60.3 days in water. Researchers from the Agricultural University of China [38] synthesized a graphene composite with a hydrogel in the formulation of which they added himexazole. First, hemexazole was added to the graphene oxide (GO) by absorption, and then dopamine was polymerized on the surface of the GO. They investigated the controlled release of pesticides. The morphology of the sample was monitored by transmission electron microscopy (TEM), where the adsorption potential occurs at 230 nm and 300 nm.

3. SYNTHETIC DYES

Various areas of modern technology make extensive use of synthetic dyes [39]. Synthetic dyes have different structures. Derivatives of anthraquinone, sulfur, triphenylmethyl (trityl), azo derivatives, phthalocyanines and indigoids are most commonly used in industry and belong to chemical classes of dyes. Azo derivatives are currently the most widely used in the industry. It is not known exactly how much paint is produced in the world in one year; however, it is estimated that about 10,000 tons are produced per year. Moreover, it is still unfamiliar how much waste dyes end up in the environment annually, except for the fact that 1-2% of the paint is released into the environment during production and 1-10% of the paint is released during use. Synthetic paints can have a great impact on environmental pollution, primarily because they have found wide applications which give rise to increased production. Due to their toxicity, they are considered extremely risky to health]40]. The release of large amounts of synthetic dyes into the environment is not only a great concern to people, but it can trigger a penalty. Contaminated paint wastewater poses a serious challenge to environmental scientists. Due to its chemical stability, it is very difficult to treat wastewater contaminated with synthetic dyes with traditional technologies. Shaul et al. [41] confirmed that out of 18 studied azo dyes 11 of them went through the process of activated sludge practically untreated, 4 (acid blue 113, red acid 151, direct purple 9 and direct purple 28) were adsorbed on activated sludge and only 3 (orange acid 7, acid orange 8 and red acid 88) are biodegradable. There are a handful of methods for treating dyes-contaminated wastewater. Adsorption has proven to be one of the most effective methods [42]. The technology of adsorption on organic or inorganic matrices has been developed as a method that is simple and at the same time economical and with the help of which pollutants can be easily removed from water [43]. In this paper, the main emphasis is on the adsorption of dyes with the help of composites of graphene (graphene oxide) and hydrogels.

Graphene hydrogel composites as adsorbents for dye pollutants

The main polluter of the environment is considered to be the rapid progress of chemical companies, but also the emission of pollutants that occur as a by-product of the process industry. The release of dyes into water bodies has been shown to interfere with the penetration of light into water. In addition, there is a slowdown in photosynthetic activity, but also through disturbances in the aquatic ecosystem [44]. Adsorption has proved to be the best method for removing paint from wastewater [45–50]. Its biggest advantages are, in addition to the simplicity of work, low cost [51]. The adsorbent is recyclable while the existence of residues is minimized. Many innovative adsorbents promising in wastewater treatment have been developed, but shortcomings such as low adsorption capacity, material fatigue and fatigue procedures still exist. Researchers are faced with the task of finding simpler and more efficient adsorbents. Akter et al. in their review paper indicate different, cellulose-based hydrogels that have found application as adsorbents in the removal of dyes from wastewater following different synthesis techniques and adsorption mechanisms [52]. Karimi et al. synthesized chitosan/κ-carrageenan, hydrogel, bioadsorbents for the removal of anionic eriochrome black-T [53]. Mahdavinia et Mosallanezhad found a green route to prepare chitosan-crosslinked κ-carrageenan bionanocomposites for removal of methylene blue [54]. Khan et al., using free-radical polymerization, synthesized green hydrogel nanocomposite which can be used as adsorbent [55]. Sivakumar et Lee removed the organic pollutant, methylene blue, from wastewater using polysaccharide-based composite hydrogels as adsorbents [56]. Due to their unique properties, graphene composites with hydrogel have attracted great interest. Tiwari et al. [57] synthesized reduced graphene oxide (RGO)-based hydrogels using the reduction of graphene oxide and sodium ascorbate. After testing and characterization, the researchers showed that RGO-based hydrogel is an excellent candidate for the adsorption of organic dyes from wastewater. They determined the toxicity of purified aqueous solutions. They came to the conclusion that the purified aqueous solutions with hydrogels based on RGO are comparable to the control experiments conducted using distilled water. Sui et al. also synthetised RGO- based hydrogels, they used simple reduction of exfoliated graphite oxide (GO) with excess vitamin C(VC) [58]. In their study, Guo et al. showed that graphene composites with hydrogel have a superior adsorption capacity when dyes are removed from the aqueous environment. They synthesized GO/polyethylenimine (PEI) hydrogels and proved that composites are efficient adsorbent for dyes. The researchers obtained the GO/PEI composite through hydrogen bonds and electrostatic interactions between PEI and GO sheets. To facilitate the gelling process, PEI is added between the GO sheets [59]. Shen et al. removed organic dyes from wastewater using the adsorption technique [60]. As an adsorbent they used hydrogel-based material composed of live Shewanella xiamenensis BC01 encapsulated within a reduced graphene oxide network. They investigate adsorption capacity with congo red and methylene blue and concluded that composite material provided universal adsorption. Composites graphene oxide-polyethylenimine-polyvinyl alcohol hydrogel (GPPH) that was synthetised by Mani and Bhandari via microwave route and then characterized, proved to be an excellent candidate for removal of cationic and anionic dyes from wastewater [61]. Graphene oxide can also be crosslinked with chitosan (CS) and carboxymethyl cellulose (CMC) for synthesized nanocomposite hydrogels. Mittal et al. synthesized these composites and used blue and anionic methyl orange dyes from contaminated wastewater, as adsorbents for removing cationic methylene [62]. Methylene blue from contaminated wastewater was also successfully removed by Liu et al. [63]. They used composites GO with hydroxypropyl cellulose.

4. CONCLUSION

As a consequence of environmental neglect and the negative effects of humans on aquatic ecosystems, people all over the world are faced with the scarcity of safe and clean water and access to drinking water. Pollution creates an increasing need for new, superior adsorbents. Composite materials of graphene and graphene oxide with hydrogels are among the materials that have found application as highly effective adsorbents of pesticides and dyes. The potential shown by graphene hydrogel composite materials as adsorbents suggests future increased production of GH composites for these purposes.

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PREČIŠĆAVANJE KONTAMINIRANIH OTPADNIH VODA UZ POMOĆ GRAFENSKIH KOMPOZITA SA HIDROGELOVIMA

Nedostatak čiste vode zahteva korišćenje novih tehnologija za prečišćavanje otpadnih voda. Adsorpcija je jednostavan i efikasan metod za uklanjanje zagađivača iz kontaminirane vode. Kompoziti grafena sa hidrogelovima našli su primenu u prečišćavanju otpadnih voda jer imaju jedinstvena svojstva kao što su porozna struktura, jedinstvena morfologija, dobra reološka svojstva, netoksičnost itd. Rad predstavlja pregled literature potencijalnih rešenja za tretman otpadnih voda korišćenjem kompozitnog materijala, grafena i grafen oksida sa hidrogelom. U ovom radu su pomenuta kompozitna jedinjenja korišćena u tretmanu različitih opasnih materija odnosno adsorpciji različitih klasa obojenih zagađivača (boja) i pesticida (organskih i neorganskih).

Ključne reči: *kontaminirana voda, pesticidi, boje, kompoziti grafen-hidrogel*