# **Polyacrylamide Soil Conditioners: The Impact on Nanostructured Clay Minerals' Aggregation and Heavy Metals' Circulation in the Soil Environment**



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### **1 Polyacrylamide Structure and Preparation**

Polyacrylamides (PAMs) are group of water-soluble, synthetic polyelectrolytes derived from acrylamide monomer in polymerization process [\[1–](#page-10-0)[3\]](#page-10-1). They are substances which dissolve, disperse, or swell in water and thus can modify physical properties of aqueous system due to thickening, gelation, emulsification, or stabilization processes. Polyacrylamide chains consist of repeating units or blocks of units that form the structure containing hydrophilic groups acting as substituents or incorporated into the macromolecule backbone. Polyacrylamides can be classified into nonionic, anionic, and cationic ones [\[4,](#page-10-2) [5\]](#page-10-3). Each of these types due to their properties is used in many industries, mainly in water treatment, mineral processing, or soil conditioning [\[5\]](#page-10-3). Application areas of abovementioned polyacrylamide types depend on the specific function of the polymer needed. The adsorption process of macromolecular compounds such as PAMs on the suspended matter particles governs the individual polymer performance. These polyelectrolytes can interact through specific or non-specific type forces with various charged substrates. The type of interaction between the polymer macromolecule and a substrate depends on the nature and polarity of the polymer itself and the surface on which it adsorbs.

Nonionic polyacrylamide has a strong hydrophilic character, greater than other nonionic, water-soluble polymers. Despite being described as nonionic synthetic

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polyacrylamides obtained as a result of polymerization or copolymerization of acrylamides, often contain in their macromolecules about 1–3% of anionic groups [\[4\]](#page-10-2). These groups are formed as a result of monomer amide groups hydrolysis. In order to obtain a polyacrylamide with a lower ionic group content, factors such as temperature, pH, monomer concentration, and type of polymerization initiator should be manipulated. Nonionic polyacrylamide can be produced from acrylamide monomer by free radical polymerization using various initiation methods. This monomer is produced by the catalytic hydration of acrylonitrile. It polymerizes with a wide range of free radical initiators but often potassium persulfate or hydrogen peroxide must be used because they allow reaction in a convenient temperature range (from 40 to 67 °C). As a result, a high-molecular-weight polymer without contaminations is obtained, which can be reduced by incorporating methanol into the reaction medium (acts as a so-called chain transfer agent):

$$
CH_2 = CH - CN \frac{\frac{H_2O}{\text{catalyst}}}{{\text{catalyst}}} CH_2 = CH - CONH_2
$$
\n
$$
Arylonitrile \qquad \text{Acrylamide}
$$
\n
$$
CH_2 = CH - CONH_2 \quad \text{initiator} \quad -(- CH_2 - CH_2 -) -
$$
\n
$$
1 \qquad \qquad COMH_2
$$
\n(2)

Acrylamide Nonionic polyacrylamide

Anionic polyacrylamide is polymer with a large number of negatively charged moieties in the long chains of the PAM macromolecules. It can be obtained by partial hydrolysis of polyacrylamide neutral amide groups, which is shown in [\(3\)](#page-1-0). This process occurs under moderate temperature by adding sodium hydroxide to the PAM solution [\[4,](#page-10-2) [6\]](#page-10-4).

$$
-(-CH_{2}-CH_{2}-) - \frac{NaOH}{N}
$$
\n
$$
-(-CH_{2}-CH_{2}) - \frac{NaOH}{N}
$$
\n
$$
-(-CH_{2}-CH_{2}-CH_{2}) - \frac{1}{N}
$$
\n
$$
-(-CH_{2}-CH_{2}) - \frac{1}{N}
$$

Polyacrylamide (PAM) Anionic PAM

<span id="page-1-0"></span>

In the case of cationic polyacrylamide, in which macromolecular chains contain positively charged groups, the basic method of preparation is the copolymerization reaction presented below:

$$
\begin{pmatrix}\n-(-CH_2 - CH -) - \\
\downarrow \\
\downarrow \\
\downarrow\n\end{pmatrix} + C_x H_y NCl \xrightarrow{\text{initiator}} \begin{pmatrix}\n-(-CH_2 - CH -) - \\
\downarrow \\
\downarrow \\
\downarrow\n\end{pmatrix} \begin{pmatrix}\n-(-CH_2 - CH -) - \\
\downarrow \\
\downarrow \\
\downarrow \\
\downarrow\n\end{pmatrix} \qquad (4)
$$

Polyacrylamide (PAM) Cationic PAM

There are four main kinds of cationic polyacrylamide synthesis technologies: in aqueous solution, in dispersion, by inverse emulsion, or through photoinitiated polymerization [\[7\]](#page-10-5). These methods had experienced numerous developments and modifications such as free radical copolymerization or grafting processes [\[8\]](#page-10-6). Due to these methods, application of PAM with relatively high molecular weight can be obtained but characterized by short duration, high prize, and poor stability. In industry, mostly free radical polymerization method including abovementioned technologies is used. The technological process of aqueous solution polymerization is presented in Fig. [1.](#page-2-0)

The aqueous solution of cationic polyacrylamide monomer is mixed with N<sub>2</sub>, initiator, i.e., 4,4'-azobis-4-cyanovaleric acid, 2,2'-azobis(2-amidinopropane)dihydrochloride and by adjusting conditions (pH, temperature, time, additives such as stabilizers, e.g., poly(acryloethyl trimethyl ammonium chloride)), the polymerization process and PAM colloids' formation are induced. This technology has many advantages (short time, safety, simplicity of method and equipment, minimal environment pollution) and for this reason is widely used in many branches.

Dispersion polymerization technology (Fig. [2\)](#page-2-1) relies on acrylamide and double bond of quaternary ammonium dissolving in the saline solution such as  $(NH_4)_2SO_4$ or NaCl and addition of initiator and stabilizer (i.e., poly(acryloylxyethyltrimethyl ammonium chloride), poly(dimethylaminoethylmethacrylate methyl chloride)) to the system. The result of this process was a discrete form of small polymer particle precipitation. These particles show unique features such as side chain structures with high positive charge density but the obtained product had low concentration and poor stability. For this reason, this method is not usually used for large technological scale in the industrial production [\[9\]](#page-10-7).

By water solution of cationic monomer and organic solvent mixing which results in the formation of water-in-oil emulsion and addition of initiator, the inverse emulsion polymerization occurs (Fig. [3\)](#page-3-0). However, besides the fact that this method allows getting high-molecular-weight polymers, it has disadvantages, i.e., the emulsification and phase separation are difficult to control [\[10\]](#page-10-8).

In the photoinitated polymerization method (Fig. [4\)](#page-3-1), the cationic polyacrylamide is obtained by photoactivation of monomer solution. This synthesis technique is characterized by many advantages: operation simplicity, easy control, low cost, and obtained product has high purity [\[11,](#page-10-9) [12\]](#page-10-10).

<span id="page-2-1"></span><span id="page-2-0"></span>

<span id="page-3-1"></span><span id="page-3-0"></span>

Polyacrylamides can also be formed by acrylamide and bis-acrylamide copolymerization initiated by free radicals. This process is vinyl addition polymerization activated by the addition of initiators. The most commonly used radical initiators are ammonium persulfate and N,N,N',N'-tetramethylethylenediamine (TEMED) or TEMED with riboflavin-5'-phosphate. Addition of TEMED catalyzes and accelerates the free radical formation rate, whereas radicals obtained from ammonium persulfate catalyze polymerization process by reaction with monomers and conversion to acrylamide free radicals. As a result of free radicals with polymer monomers reaction, they are transformed into further free radicals reacting with inactive monomers which eventually starts the polymerization reaction [\[13](#page-10-11)[–15\]](#page-11-0).

#### **2 PAM Impact on Soil Aggregation**

Particulate suspensions, including clay minerals, are used in many industrial processes that result in high-quality products (e.g., ceramics, paper coatings, cosmetics, paints, rubbers, and other plastics). These suspensions are usually highly concentrated and for this reason the maintenance of a system where solid particles are well dispersed is significantly difficult. Thus, the big importance is control of colloidal properties and dispersion stability [\[15,](#page-11-0) [16\]](#page-11-1).

The addition of a high molecular compound affects the stability of colloidal suspensions and its rheological properties. Depending on the conditions, the presence of polymer in the system may increase or decrease aggregation stability. The protective effect of the polymer resulting in an increase in the suspension stability is called polymer stabilization, whereas that causing a decrease in this parameter—polymer flocculation. Polymer stabilization or flocculation is the result of a macromolecular compound adsorption on the surface of particles of the dispersed phase or the presence of nonadsorbed polymer macromolecules in the dispersive phase [\[17\]](#page-11-2). Due to the large size of the macromolecular compounds and the presence of many active sites in their chains, polymer shows a much greater tendency to adsorption than compounds with a lower molecular weight when contacting the surface of a solid. In addition, the polymer macromolecule can adsorb onto the solid surface in a variety of ways, resulting in a number of conformations such as trains, loops, and tails (Fig. [5\)](#page-4-0) [\[6,](#page-10-4) [13,](#page-10-11) [17–](#page-11-2)[20\]](#page-11-3).



<span id="page-4-0"></span>**Fig. 5** Conformations of adsorbed polymer on the solid surface [\[13\]](#page-10-11)

The concentration of the adsorbing polymer affects its ability to stabilize the colloidal system. At high concentrations, the solid surface is completely covered by the polymeric layer. When the polymer-coated particles approach each other, the interpenetration of the polymer layers occurs. Consequently, the conformational entropy of the polymer segments decreases, whereas the free enthalpy increases. As a result, the repulsion (steric stabilization) between the particles with the adsorbed polymer is observed. In the case when the adsorbing polymer is a polyelectrolyte, steric stabilization can be combined with electrostatic stabilization and electrosteric stabilization takes place in the system (also as a result of repulsion of adsorption layers with an identical charge) [\[6\]](#page-10-4).

The polymer adsorbed on the solid surface can form loops and tails, which means that only a few fragments of polymer chains have direct contact with the solid surface. The addition of some amount of polymer to the system that will not provide complete coverage of the surface contributes to the formation of polymer bridges. This is only possible if the range of electrostatic interaction between the particles is smaller than the length of the polymer loops and tails. As a result of the bridging flocculation process occurs, i.e., formed aggregates size until they are separated from the solution in the form of a precipitate or a turbid suspension. Flocculation can also be a result of the solid charge neutralization by adsorbed polyelectrolyte macromolecules with an opposite charge [\[21–](#page-11-4)[23\]](#page-11-5).

The phenomenon of stabilization and flocculation of the dispersed systems through the addition of a polymer is widely applied in many industries [\[6,](#page-10-4) [21\]](#page-11-4). Adsorption of high molecular compounds is used mainly in the wastewater treatment [\[24,](#page-11-6) [25\]](#page-11-7), in agriculture [\[26](#page-11-8)[–28\]](#page-11-9), chemical industry (production of paints, inks, varnishes, plastics) [\[29,](#page-11-10) [30\]](#page-11-11), paper industry [\[31,](#page-11-12) [32\]](#page-11-13), in the production of cosmetic preparations [\[33\]](#page-11-14), food industry [\[34](#page-11-15)[–36\]](#page-12-0), ceramics [\[37,](#page-12-1) [38\]](#page-12-2), and medicine and pharmacy [\[39](#page-12-3)[–41\]](#page-12-4). Polymer adsorption is also used in soil stabilization and conditioning [\[28,](#page-11-9) [42](#page-12-5)[–46\]](#page-12-6). Due to the high intermolecular bond strength, the polymer ensures an additive controlling of soil erosion, which is safe for health and the environment. It strengthens soil cohesion by binding loose mineral particles [\[47\]](#page-12-7). In addition, it

affects the reduction of sludge transport; thus, the soil erosion process is limited [\[47–](#page-12-7) [49\]](#page-12-8). The macromolecular compounds interact mainly with clay minerals found in soils or sediments. Adsorbing on the surface of the grains, they affect their properties. Changing the surface properties of a mineral, soil, or sediment, they affect the fate of other compounds, i.e., nutrients, heavy metals, and organic substances present in the surrounding environment [\[50\]](#page-12-9).

However, from a technical point of view, it is necessary to study the concentrated systems of solid particles. The main problem of this type of system is maintaining adequate dispersion or achieving adequate rheological properties. It is already known that concentrated suspensions are stabilized or flocculated by the addition of a polymer, but the role of conformation of the macromolecular compound in these phenomena remains unclear. This is mainly due to difficulties in measuring adsorption and determining the conformation of the polymer substance in highdensity systems. However, there are several techniques that allow such research. These are Fourier-transformed infrared spectroscopy (FT-IR), nuclear magnetic resonance (NMR), electron spin resonance (ESR), and fluorescence spectroscopy. Using the fluorescence spectroscopy and ESR technique, the conformation of poly(acrylic acid) on the solid surface and its effect on the flocculation process was determined. Based on the obtained results, it was found that the higher the molecular weight of the polymer is, the greater the degree of coiling of its macromolecules is. The more developed conformation of the high molecular compound is synonymous with the formation of a larger number of tails and loops on the surface of the solid, which in turn increases the efficiency of the flocculation process (creation of more numerous bridges) [\[13,](#page-10-11) [51\]](#page-12-10). It is also worth mentioning that the research on the impact of the polyacrylamide molecular weight on the adsorption, and flocculation of suspension process is also important. The higher the molecular weight of the polymer is, the greater the adsorption capacity is observed. A polymeric substance with a higher molecular weight is a much better flocculant, because a larger amount of it is adsorbed on the solid surface. In addition, the conformation of polymer chains has also been shown to depend on their molecular weight, which is manifested in an increase in chain length. This promotes the development of polymer macromolecules conformation and, as a consequence, flocculation efficiency by bridging increases.

Adsorption of polymers such as polyacrylamide and their role in the process of stabilization and flocculation of dispersed systems has a great scientific interest [\[52–](#page-12-11)[61\]](#page-13-0). The type of action of the macromolecular compound, whether it acts as a stabilizer or flocculant, is influenced by many factors such as structure, conformation, solubility, and degree of polymerization [\[58,](#page-13-1) [60\]](#page-13-2), as well as the energy of interaction of polymer chains with a solid surface [\[62\]](#page-13-3). Understanding the mechanism of polymer adsorption is important considering the production of increasingly effective dispersants. So far, many studies on adsorption and the influence of the high molecular compound presence on the interaction between solid particles have been presented. These studies concerned: the influence of pH, ionic strength, and the polymer charge on the adsorbed amount of the macromolecular compound [\[46,](#page-12-6) [52,](#page-12-11) [63–](#page-13-4)[68\]](#page-13-5); kinetics of the flocculation process [\[54\]](#page-13-6); and the role of polymer conformational changes.

Currently, many studies refer to the assessment of mineral particle stability in colloidal systems, the nature of interactions between them, as well as the impact of high molecular compound presence on the suspension stability. These studies mainly refer to polyacrylamide (PAM) which is a non-toxic, environmentally friendly polymer that has the ability to stabilize and improve soil structure [\[69,](#page-13-7) [70\]](#page-13-8). Its beneficial effect is related to maintaining or increasing soil aggregation and pore continuity [\[71–](#page-13-9)[73\]](#page-13-10), increasing aggregate stability, and thus soil structural stability [\[74,](#page-14-0) [75\]](#page-14-1). The potential benefits of using PAM are influenced by a number of relationships between the properties of the polymer (its molecular weight, type, amount of surface charge, conformation of macromolecules, and density of the charge) and the properties of the soil (its type, texture, organic matter and clay mineral content, ionic strength, pH value, composition, and ion concentration in soil solution) [\[1,](#page-10-0) [76–](#page-14-2)[79\]](#page-14-3). The type of polymer (and thus charge present in macromolecules) affects the amount of its adsorption on solid surface. The polymer adsorption increases with the following order: anionic PAM < nonionic PAM < cationic PAM [\[1\]](#page-10-0). Nonionic polyacrylamide adsorption on clay minerals is mainly caused by van der Waals forces [\[80\]](#page-14-4), whereas cationic or anionic forms adsorb and bind soil particles through electrostatic forces or bridge formation between PAM charged moieties and solid surface group [\[1,](#page-10-0) [80\]](#page-14-4). Malik and Letey [\[81\]](#page-14-5) concluded that due to differences in polymer chain conformations influenced by ionic PAM hydrolysis degree the stabilization of soil by polyacrylamide can vary. Adsorbed polymeric chain conformation results from solution pH value and surface charge. The higher the percent of hydrolysis (greater amount of positively or negatively charged groups) was, the more linear, well-developed macromolecular chain and more extended conformation was obtained, and thus greater adsorption was observed [\[81,](#page-14-5) [82\]](#page-14-6). Adsorption of PAM characterized by 2 and 20% hydrolysis degree was studied [\[83\]](#page-14-7). The adsorption of polymer with greater amount of hydrolyzed groups was higher, whereas the more significant stability of soil aggregates with PAM 2% was observed. Several papers related to the effect of electrolyte present on polymer adsorption and its ability to soil stabilizing. The electrolyte presence in adsorption system enhances the effectiveness of soil aggregate stabilization by polymer and its adsorption on mineral surface [\[83\]](#page-14-7).

The addition of polymer to the soil stabilizes existing aggregates and strengthens the mutual binding of adjacent particles. The stabilizing efficiency of polyacrylamide is mainly determined by the adsorption of its macromolecules on the surface of soil particles [\[84\]](#page-14-8). The aggregate formation contributes to several beneficial phenomena such as stability increase, reinforcement of soil structure, water infiltration promotion as well as erosion limitation [\[86\]](#page-14-9). Shainberg et al. [\[85\]](#page-14-10) and Ben-Hur et al. [\[86\]](#page-14-9) conducted infiltration studies with polyacrylamide and soils rich in clay mineral fractions. Application of PAM flocculant promotes surface sealing prevention which results in greater infiltration rate. Fox and Bryan [\[87\]](#page-14-11), Stern et al. [\[68\]](#page-13-5), and Smith et al. [\[88\]](#page-14-12) determined the influence of PAM presence on soil erosion and field runoff reduction. Soils treated with polyacrylamide maintain their original physical state in water erosion conditions. By stabilization of clay mineral aggregates by polymer adsorption, the water infiltration rate improvement and soil erosion reduction are observed.

The PAM adsorbed amount may also depend on the soil and clay mineralogy [\[1,](#page-10-0) [76,](#page-14-2) [82\]](#page-14-6). The amount of adsorbed polymer on the surface of soil particles is up to three orders of magnitude smaller than the amount adsorbed on clay mineral particles. Under neutral or acidic pH, the adsorption of anionic polyacrylamide on the surface of clay materials such as montmorillonite, kaolinite, and illite is comparable. However, under basic conditions, the amount of adsorbed polymer decreases in order: illite, kaolinite, and montmorillonite [\[46,](#page-12-6) [76\]](#page-14-2). What is more, under alkaline pH conditions greater adsorption of anionic PAM on the illite surface compared to montmorillonite one was observed. The presence of polyacrylamide with high molecular weight contributes to flocculation process. However, the structure of flocs depends on clay mineral type. These micro-aggregates of soil particles with polyelectrolyte can be formed in both neutral and acidic solutions. Other studies focused on the stability of smectic soil aggregates in the presence of PAM [\[74\]](#page-14-0). It turned out that the macromolecules that entered the pores of the aggregates did not have a significant effect on the stability of their structure. On the other hand, this polymer increased the percentage of stable aggregates in kaolinite soils with different structures [\[89\]](#page-14-13) and particle size [\[90\]](#page-14-14). The formation of aggregates, stability of macro- and micro-aggregates, their degradation mechanism, and chemical dispersion depend not only on the amount of clay minerals and organic matter but also on soil mineralogy [\[91\]](#page-14-15). The influence of the mineralogical composition is difficult to determine because soils usually contain a mixture of clay materials and their properties are modified by binding with other minerals. Under unstable conditions (such as high sodium content or low electrolyte concentrations), soils with high montmorillonite content are unstable, soils with high kaolinite content are relatively stable, and soils rich in 2:1 clay minerals or with low content of montmorillonite show intermediate stability [\[92,](#page-14-16) [93\]](#page-14-17). The effectiveness of PAM as a soil aggregate stabilizing factor has also been studied. The addition of polymer had a positive effect on increasing the moisture content of soil aggregates, thanks to which their stability improved (in ascending order: kaolin < illitic < smectite soils).

The content of clay in the soil and their mineralogy significantly affects the stabilization of aggregates by PAM. The effectiveness of stabilization may result from the natural stability of aggregates and soil properties affecting polymer adsorption on the solid surface. The optimal pH value and the presence of easily soluble minerals, which provide divalent cations to the soil solution, can contribute to increasing PAM adsorption on the surface of soil particles and the ability to increase aggregate stability.

Polyacrylamide treatments has significant impact on crop growth and yield as well as nutrient effect [\[28,](#page-11-9) [94–](#page-15-0)[97\]](#page-15-1). Wallace [\[98\]](#page-15-2) studied the interaction of polyacrylamide with the following nutrients: P, Zn, Cu, Fe, Mn, and Mo. The concentration of these elements in the soil solution containing PAM is significantly different than in systems without macromolecular additive. Moreover, these differences in microelements concentration affect the plant growth—the studies shown that plant growth is improved in the nutrient solutions with polyacrylamide soil flocculant [\[98\]](#page-15-2).

## **3 PAM Effect on Heavy Metals' Accumulation in the Soil Environment**

Clays and modified clays have been found particularly useful for adsorption of heavy metals' ions and are the objects of several experimental works [\[99–](#page-15-3)[114\]](#page-16-0). Clays mineral due to their internal structure can adsorb metals such as As, Cd, Cr, Co, Cu, Fe, Pb, Mn, Ni, and Zn from aqueous medium. However, their adsorption capacity depends on the type of clay used and also differs for various metals [\[99\]](#page-15-3). In many papers, the limiting or enhancing conditions' effect on the adsorbent efficiency of the clay materials was studied. The influence of pH value, temperature, quantitative efficiency of the individual mineral, modifications of the clay, and its composites in removing various contaminants were examined.

Investigations of pH, contact time, temperature, and initial metal cation concentration influence on uranium(VI) ions' adsorption on bentonite clay were carried out [\[103\]](#page-15-4). Aytas et al. [\[103\]](#page-15-4) showed that the mineral adsorption properties can change due to temperature and time increase—the highest capacity was observed in the case of bentonite calcinated under 400 °C and at the beginning of adsorption process.

Mishra and Patel [\[104\]](#page-15-5) studied lead and zinc ions' removal from water by various adsorbents, i.e., kaolin and bentonite depending on the time, pH, adsorbent dosage, and contact time. The obtained results showed no significant differences in heavy metals adsorption in various pH values. The highest Pb and Zn removal from solution was observed when an increase in adsorbent dosage occurred. The increase of ionic strength of electrolyte results in decrease of metal ion adsorption.

The removal of Pb, Cd, Ni, and Cu ions from aqueous solution by kaolinite was studied by Jiang et al. [\[111\]](#page-15-6). The adsorption was carried out in different conditions differing with initial metal ion concentration, pH, ionic strength of electrolyte, and contact time. Obtained results showed that the pH value of solution had most significant impact on heavy metal adsorption on clay surface, and adsorption equilibrium was obtained after 30 min. With the increasing metal concentration, its adsorption also increases (due to stronger driving forces to mineral surface).

Bhattachryya and Sen Gupta [\[100\]](#page-15-7) reviewed the removal of toxic metal ions by natural kaolinite, montmorillonite, and their modified forms by pillaring with polyoxy cations such as  $Zr^{4+}$ ,  $Al^{3+}$ ,  $Si^{4+}$ ,  $Ti^{4+}$ ,  $Fe^{3+}$ ,  $Cr^{3+}$ , and  $Ga^{3+}$ . Comparison of adsorption capacity of these adsorbents indicated that the natural and modified montmorillonite may adsorb much more heavy metals than kaolinite sorbents. Various metal ions can be captured from solution by clay mineral adsorbent and their modified forms [\[99\]](#page-15-3). The sorption affinity to As anions improvement by pre-treatment of kaolin or bentonite minerals with Fe(II), Fe(III), Al(III), and Mn(II) ions was shown [\[107\]](#page-15-8). Na et al. [\[109\]](#page-15-9) demonstrated that by pillaring Ti to montmorillonite, the arsenate or arsenite removal efficiency from aqueous solution can be increased. The adsorption of As ions as a function of pH value, contact time, temperature, coexisting ions' presence, and ionic strength was studied. Comparing all examined ions (phosphate,

nitrate, and sulfate), only phosphate one had noticeable effect on heavy metal adsorption. The decrease in temperature caused an increase in As-adsorbed amount on clay surface.

Oliveira et al. [\[101\]](#page-15-10) compared nickel, cadmium, zinc, and copper cations' adsorption on bentonite clay and iron clay minerals with magnetite composite from aqueous solution. It was shown that the presence of iron oxide in adsorbent structure enhanced the adsorption capacity of bentonite. Yuan et al. [\[105\]](#page-15-11) investigated the removal of Cr(VI) by montmorillonite supported by magnetite nanoparticles. The clay mineral modified by nanoparticles of magnetite showed greater adsorption capacity per unit mass. Moreover, the chromium adsorption process was highly pH-dependent.

Several papers related to the adsorption of heavy metals on mineral soil surface modified by polymeric substances. Gecol et al. [\[102\]](#page-15-12) compared tungsten removal efficiency by natural and chitosan-coated montmorillonite. The effects of W concentration as well as pH value were studied. The W-adsorbed amount increased with decrease in solution pH value and metal concentration. However, clay mineral coated with biopolymer was found to be much more effective adsorbent compared to natural montmorillonite.

Wiśniewska et al. [\[115,](#page-16-1) [116\]](#page-16-2) and Fijałkowska et al. [\[117,](#page-16-3) [118\]](#page-16-4) examined the anionic and cationic PAM adsorption on clay minerals, i.e., montmorillonite and kaolinite and its impact on the heavy metal ions' accumulation. Both polyacrylamides may strengthen the Pb(II) and Cr(VI) ions' adsorption on the aluminosilicate surface.

Pandley and Mishra [\[106\]](#page-15-13) used chitosan/clay nanocomposite for Cr(VI) ions' removal from aqueous solution. The adsorption of chromium(VI) ions on montmorillonite surface modified with biopolymer was carried out in different pH values. However, the highest metal ions' adsorbed amount was observed at pH 3.

Another study concerned selenium adsorption on chitosan–montmorillonite composite [\[108\]](#page-15-14). The adsorption process on the composite material was pHindependent. It has been shown that clay mineral modified by chitosan is a highperformance adsorbent which allows removal of the metal ions with a high concentration from solution. The  $Cu(II)$  ions' adsorption process on bentonite surface with polyacrylamide gel in the function of pH, ionic strength, adsorbent content, metal ion concentration, and temperature was studied [\[110\]](#page-15-15). Obtained results indicated high pH, ionic strength, and temperature dependency of copper ions' sorption process. With decrease in temperature and ionic strength, the adsorbed amount of  $Cu(II)$  ions increased.

The effects of contact time, adsorbent dosage, and pH of the initial solution on the  $Hg^{2+}$  removal by polyacrylamide/attapulgite were studied [\[113\]](#page-15-16). The mercury ions' adsorption on PAM–attapulgite surface increases with increasing contact time and pH of the initial suspension but decreases with clay mineral composite dosage. Zhou et al.  $[112]$  studied Hg(II), Pb(II), and Co(II) ions' adsorption on polyacrylamide/attapulgite system. The obtained results showed that the clay mineral adsorbent modified by PAM exhibits the highest sorption capacity and selectivity in relation to mercury ions, which is manifested in their greatest adsorption compared to other ions.

Gu et al. [\[114\]](#page-16-0) studied selective heavy metal adsorption on unmodified mineral and polyacrylamide–vermiculite composite surfaces. The adsorbent modified by polymer showed much greater efficiency in Pb(II) adsorption at different pH values than unmodified one and better selectivity for  $Pb(II)$  ion in comparison to  $Zn(II)$ ,  $Cd(II)$ , and Cu(II) ions.

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