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Research Paper

## An overview of geological originated materials as a trend for adsorption in wastewater treatment

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### ABSTRACT

Adsorption is a unit operation widely used for the tertiary treatment of the most diverse effluents, whose mechanism is based on removing recalcitrant compounds from the organic and inorganic origin. In this process, choosing a suitable adsorbent is a fundamental point. This review article focuses on the adsorbents with natural geological origin: minerals, clays, geopolymers, and even wastes resulted from mining activity. Therefore, over 450 articles and research papers were explored. These materials' main sources are described, and their characteristics, composition, and intrinsic properties are related to adsorption. Herein, we discuss the effects of several process parameters, such as pH, temperature, pollutant, and adsorbent concentration. Furthermore, equilibrium, kinetics, and thermodynamic aspects are also addressed, and relevant regeneration prospects and final disposal. Finally, some suggestions and perspectives on applying these adsorbents in wastewater treatment are presented as future trends.

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### 1. Introduction

Around 95% of the Earth's crust is constituted by rock and rock-based composites; therefore, geological materials are abundant and accessible (Banning, 2020; Ronov and Yaroshevsky, 1922; Taylor and McLennan, 1985). Meanwhile, due to our planet's population's growth and need for more significant development, more and more areas are explored, and more cities are built. Because of these anthropogenic actions, geological materials are dislocated and disposed of as residues, creating an environmental problem (Bettencourt et al., 2007; Folke et al., 2020; Ghernaout et al., 2020). After considering that these materials are mostly made up of silicon, oxygen, and other metal oxides (Folk, 1954), several studies have demonstrated interest in applying and investigating their natural properties towards adsorption, aiming the removal of environmental pollutants as a bright and elegant way of solving two growing problems (Atun et al., 2009; Bundschuh et al.,

2011; Dong et al., 2020; Gu et al., 2019; Li and Arai, 2020; Roy and Krapac, 1994; Teymouri et al., 2020b; Tsai et al., 2006).

Adsorption is one of the most effective techniques in advanced wastewater treatment and can be employed to reduce or even remove hazardous organic and inorganic pollutants present in the effluent (Burakov et al., 2018; Crini, 2006; Dabrowski, 2001; De Gisi et al., 2016; Dotto et al., 2016; Dotto and McKay, 2020; Lütke et al., 2019). The process concerns the accumulation of a substance at the interface between two phases, liquid-solid or gas-solid (Liu and Jiang, 2010; Tóth, 1995). The substance that accumulates at the interface is named adsorbate, while the solid on which adsorption occurs is the adsorbent (Fig. 1) (Dotto and McKay, 2020).

Adsorption can occur by chemical sorption and physical sorption (Di Toro, 1985; Hammond and Conner, 2013). Chemical adsorption, or chemisorption, consists of forming strong chemical associations – or even bonds – between molecules or ions of adsorbate to the adsorbent surface (Breeuwsma and Lyklema, 1973; Fan et al., 2020; Hong et al., 2020). Contrarily, physical adsorption, or physisorption, is described by intermolecular interactions between adsorbate and adsorbent. Nevertheless, the main physical forces controlling adsorption are van der Waals forces, hydrogen bonds, polarity, dipole-dipole, and  $\pi$ - $\pi$  interactions (Chen et al., 2020; Halsey, 1948; Li et al., 2019a).

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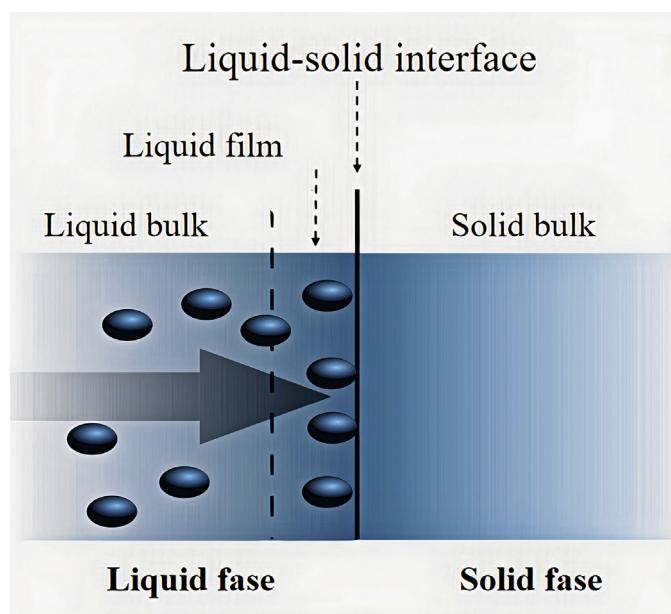


Fig. 1. Representation of the adsorption mechanism in liquid and solid faze.

Overall, adsorption provides an attractive alternative for the treatment of polluted waters, especially if the sorbent is inexpensive, which geological originated materials can be (Asfaram et al., 2014; Ghaedi et al., 2015; Saif Ur Rehman et al., 2013; Shams et al., 2013a; Uddin, 2017). This review article presents how natural, geological originated materials, such as minerals, clays, geopolymers, and even waste resulting from mining activity, can be applied to water treatment and decontamination purposes. The geological materials characteristics, composition, and intrinsic properties related to adsorption are presented. The effects of several process parameters, such as pH, temperature, pollutant, and adsorbent concentration, are discussed. Furthermore, equilibrium, kinetics, and thermodynamic aspects are also addressed mainly, according to each application. Finally, some suggestions and perspectives on applying these adsorbents in wastewater treatment are presented as future trends.

## 2. Mineral adsorbents

More than 4000 naturally occurring minerals have been found on Earth (Haldar and Tišljarić, 2013). To be classified as a mineral, the solid

must be inorganic with characteristic chemical composition and specific crystal structure (Haldar and Tišljarić, 2013; Mitchell et al., 1994). Due to the abundant presence of oxygen, silicon, and aluminum, silicates and aluminosilicates are quantitatively the most relevant class of minerals. Furthermore, the following classes are recognized: native elements; sulfide, telluride, arsenide, and selenide minerals including sulfosalts of antimony and bismuth; halides; oxides; hydroxides; carbonates; nitrates; borates; chromates; tungstates; molybdates; phosphates; arsenates; vanadates; silicates and aluminosilicates (Mitchell et al., 1994). For adsorption application purposes, the most relevant minerals are silicates, hydrous silicates, oxides, and carbonates (Cornell, 1993; Pehlivan et al., 2009; Rao et al., 1991; Zhang et al., 2019b).

In general, there are several critical parameters for a material to be considered a potentially promising adsorbent, such as its surface area, and porosity, surface charge, mechanical strength, and the surface chemistry of the adsorbent (Dotto et al., 2016; Lütke et al., 2019). In Table 1, it is presented the classifications and the average surface area, connecting geological materials and adsorption (Brantley and Mellott, 2000; Giménez et al., 2007; Hodson, 1998; Meier et al., 1994; Nandi et al., 2009; Reddy and Claassen, 1994; Sorwat et al., 2020; Walker et al., 2003; Wang et al., 2020a; Wang and Zhang, 2020; Yan et al., 2020). Several examples of the application of minerals in adsorption have been reported for various types of contaminants. Talc, chalcocopyrite, and barite were tested against the removal of lead ions from liquid wastes (Rashed, 2001), while bisphenol-A adsorption onto andesite, diatomaceous earth, titanium dioxide, and activated bleaching earth were studied. Their results were compared to those of different carbon-based materials (Tsai et al., 2006). Bituminous coal, pumice stone, coconut coal, and volcanic zeolite were evaluated to remove nitrogen, phosphorus, aluminum, iron, cyanobacteria, and saxitoxins (Guimarães Neto and Aguiar, 2020). Furthermore, mineral adsorbents' addition to porous concrete was also investigated to reduce groundwater pollution (Teymouri et al., 2020a,b).

### 2.1. Silicates

#### 2.1.1. Feldspar

Feldspar is, up to now, little explored when compared to other minerals. In a thorough search, very few related studies were found on its adsorption capacity towards environmental pollutants. However, feldspar is estimated to be the greatest rock-forming mineral in the earth crust (Branlund and Hofmeister, 2012; Horai, 1971; Janecke and Evans, 1988); hence, it is a potentially low-cost industrial material that can be employed in environmental, economic processes (Yazdani et al., 2012). They can also undergo thermal treatment operations to improve its sorption capacity (Pelte et al., 2000).

Table 1

Mineralogical classification of geological originated adsorbents reported in the literature.

Group	Mineral	Molecular formula	Average surface area ( $\text{m}^2 \text{g}^{-1}$ )	Reference
Silicates	Feldspars	$\text{KAlSi}_3\text{O}_8$	0.8	Hodson (1998)
		$\text{NaAl}_2\text{Si}_3\text{O}_8$		
		$\text{CaAl}_2\text{Si}_2\text{O}_8$		
	Micas	$\text{KAl}_2(\text{Si}_3\text{Al}) \cdot \text{O}_{10}(\text{OH})_2$	100	Meier et al. (1994)
	Amphiboles	$\text{K}_2(\text{MgFe})_6(\text{SiAl})_8\text{O}_{20}(\text{OH})_4$	3.1	Brantley and Mellott (2000)
Hydrous silicate	Pyroxenes	Ca, Mg, Fe, Al silicate	2.3	Reddy and Claassen (1994)
	Olivine	$(\text{MgFe})_2\text{SiO}_4$	1.5	Brantley and Mellott (2000)
	Kaolinite	$\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$	13.69	Meier et al. (1994)
	Quartz	$\text{SiO}_2$	0.17	Sorwat et al. (2020)
Oxide	Aluminum	$\text{Al}_2\text{O}_3$	122	Wang et al. (2020a)
	Iron oxides	$\text{Fe}_2\text{O}_3$	0.381	Giménez et al. (2007)
		$(\text{Fe}_2\text{O}_3)_2(\text{H}_2\text{O})_3$	10.26	Yan et al. (2020)
	$\text{Fe}_3\text{O}_4$	37.21	Sorwat et al. (2020)	
Carbonates	Calcite	$\text{CaCO}_3$	0.5	Wang and Zhang et al. (2020)
	Dolomite	$\text{CaMg}(\text{CO}_3)_2$	0.7	Yan et al. (2020)

The adsorption isotherms, kinetics, and thermodynamics of the cationic dyes Basic red 18 and Basic Blue 41 onto feldspar were investigated, considering both single and binary systems. The optimal adsorbent dosage was  $2.5 \text{ g L}^{-1}$ , with the dye's concentration ranging from  $12.5$  to  $50 \text{ mg L}^{-1}$  (Yazdani et al., 2012). The dye uptake capacity increased as pH increased from 2.5 to 8. This behavior is an exciting point because most effluents' final pH is around 5–8; therefore, no pH adjustment would be necessary (Boczkaj and Fernandes, 2017). Their adsorption behavior followed Langmuir for single and extended Langmuir isotherms for binary systems, respectively. The adsorption kinetics was found to conform to pseudo-second-order kinetic for both systems. Meanwhile, the thermodynamic data showed that dye adsorption onto Feldspar was spontaneous, endothermic, and physisorption was the dominant mechanism (Yazdani et al., 2012).

Besides organic molecules, the adsorption of potentially harmful ions has also been studied in natural and modified feldspar. Ferric ions, uranium (U(VI)), and arsenic (As(V)) are some examples (Al-Anber, 2015; Ding et al., 2014; Yazdani et al., 2016). Natural feldspar was useful for removing  $\text{Fe}^{3+}$  ions from an aqueous solution. The maximum removal was found at a low-level initial concentration of  $\text{Fe}^{3+}$  ions ( $30 \text{ mg L}^{-1}$ ) on  $40 \text{ g L}^{-1}$  of adsorbent dosage,  $30^\circ\text{C}$ , and a contact time of 180 min. Finally, it was concluded that the adsorption mechanism was chemisorption (Al-Anber, 2015). In Table 2, a summarization of the technical results on feldspar adsorption is presented.

### 2.1.2. Micas

Micas are monoclinic pseudo-hexagonal crystals, similar in chemical composition, with the general molecular formula of  $\text{X}_2\text{Y}_{4-6}\text{Z}_8\text{O}_{20}(\text{OH}, \text{F})_4$  – where X is K, Na, or Ca, Y is Al, Mg, or Fe, and Z is Si or Al but also may include  $\text{Fe}^{3+}$  or Ti (Frye, 2006). Some examples are biotite, muscovite, lepidolite, phlogopite, zinnwaldite, and margarite (Deer et al., 2013). This mineral was employed in few studies, mostly for the adsorption of metal ions, surfactants, and pharmaceuticals (Alexandre-Franco et al., 2011; Gier and Johns, 2000; Klapayta et al., 2001; Li et al., 2019b, 2019c; Martín et al., 2018, 2019; Orta et al., 2018; Osuna et al., 2019; Pazos et al., 2017).

### 2.2. Oxides

The oxides include about 40 cationic elements in their composition, such as Si, Ti, Nb, Ta, Mn, Al, Mg, Sn, and Zr combined with oxygen or hydroxyl (Frye, 2006; Marfunin, 1994). The most commonly known and occurring are cuprite ( $\text{Cu}_2\text{O}$ ), corundum ( $\text{Al}_2\text{O}_3$ ), hematite ( $\text{Fe}_2\text{O}_3$ ), cassiterite ( $\text{SnO}_2$ ), magnetite ( $\text{Fe}_3\text{O}_4$ ), ilmenite ( $\text{FeTiO}_3$ ), perovskite ( $\text{CaTiO}_3$ ), brucite ( $\text{Mg}(\text{OH})_2$ ) and gibbsite ( $\text{Al}(\text{OH})_3$ ) (Frye, 2006; Marfunin, 1994). Among the above-cited minerals, magnetite, hematite, and ilmenite stand out. Because iron is present in their structure, simultaneous degradation reactions can occur, a process known and well

established as Fenton reactions (Gheraout et al., 2020). Moreover, the naturally occurring oxide of titanium,  $\text{TiO}_2$ , is typically obtained from the minerals ilmenite, rutile, and anatase (Lahiri and Jha, 2007). It is widely used as a pigment in paints and personal care products, and its activity as a photocatalyst is well-established (Diebold, 2003; Weir et al., 2012). Still, these light-interacting properties are mostly used to degrade pollutants, not adsorption per se (Shiraishi et al., 2005).

Magnetic properties are of most interest, considering the difficulty of separating solid catalysts from aqueous solutions (Serpone et al., 2010). Therefore,  $\text{Fe}_3\text{O}_4$  is the most popular iron oxide: there are about 15 times more studies published on its regard than hematite and ilmenite. Magnetite properties as adsorbent are widely known and accepted (Milonjić et al., 1983; Udovic and Dumesic, 1984), mostly by the adsorption of ions as phosphate, arsenite, and arsenate (Daou et al., 2007; Yean et al., 2005), but its use for adsorption of organic molecules has been reported only with hybrid materials (Cheng et al., 2012b; Qin et al., 2014; Vieira et al., 2020).

Overall, this class of natural geological materials is interesting for adsorption applications because, in their structure, they have metal atoms electrostatically charged or in different oxidation states, which enhances the attraction of the pollutant to the adsorbent surface and, consequently, increases the total removal efficiency (Khaleel et al., 1999; Velazquez-Jimenez et al., 2015; Wang et al., 2020a). Table 3 is presented a portrait of the most relevant studies on adsorption onto oxides.

### 2.3. Carbonates

In geology and mineralogy, minerals dominated by the carbonate ion,  $\text{CO}_3^{2-}$ , are designated simply as carbonates, which are incredibly varied and omnipresent in chemically precipitated sedimentary rocks (Lippmann, 1973). The trivial are calcite, dolomite, and siderite (Curtis and Coleman, 1986; Pearce et al., 2013). Calcite,  $\text{CaCO}_3$ , is the primary constituent of limestone and the main component of mollusk shells and coral skeletons (Lorens, 1981; Somasundaran and Agar, 1967; Wray and Daniels, 1957). Dolomite,  $\text{CaMg}(\text{CO}_3)_2$ , is used as an ornamental stone, a concrete aggregate, and a source of magnesium oxide (Baker and Kastner, 1981; Sibley and Gregg, 1987; Warren, 2000). Siderite,  $\text{FeCO}_3$ , is the essential iron ore and has been used for steel production (Matthiesen et al., 2003; Mozley, 1989; Zhu et al., 2020). Moreover, some other popular carbonates, such as sodium carbonate and potassium carbonate, have been used since antiquity for cleaning and preservation, as well as for the manufacture of glass (Freilich et al., 2014; Konkol and Rasmussen, 2015; Krumbein et al., 1991; Wood et al., 1984). Carbonates are widely used in industry, e.g., in iron smelting, as a raw material for Portland cement and lime manufacture, in the composition of ceramic glazes, and more (Al-Shalabi and Sepehrnoori, 2016; Boyjoo et al., 2014; Clements, 2003; Dhami et al., 2013).

**Table 2**  
Main feldspar applications in adsorption.

Adsorbent	Dosage ( $\text{g L}^{-1}$ )	Adsorbate	$C_0$ ( $\text{mg L}^{-1}$ )	$q_{\text{max}}$ ( $\text{mg g}^{-1}$ )	Reference
Natural feldspar	2.5	Basic blue 41	25	11.11	Yazdani et al., 2012
Surfactant-modified feldspar	0.5	Basic red 18	25	10.75	Yazdani et al., 2012
		Acid black 1	10	6.37	
		Acid red 14	10	3.98	
		Acid black 1	10	3.98	
Chitosan/Feldspar biohybrid	4	Acid black 1	25	4.16	Yazdani et al., 2014
			50	8.33	
			75	12.5	
			100	16.12	
			125	17.24	
			125	17.24	
Natural feldspar	40	$\text{Fe}^{3+}$	30	25	Al-Anber, 2015
Raw sodium feldspar	1	U(VI)	0.5	0.314	Ding et al., 2014
Natural feldspar	8		5	235.3	Yazdani et al., 2016

**Table 3**  
Most important aspects of the adsorption studies using oxides for pollutant removal.

N.	Adsorbent		Adsorption test							Reference
	Type	Specific surface area (m <sup>2</sup> g <sup>-1</sup> )	Pollutant	Adsorbent dosage	Pollutant initial concentration C <sub>0</sub>	pH	Amount adsorbed q <sub>max</sub>	Isotherm type	R <sup>2</sup>	
1	Cuprite	2.15	Diethyldithiocarbamate	20 g L <sup>-1</sup>	30 g L <sup>-1</sup>	7	6.08 g g <sup>-1</sup>	-	-	
2	Perovskite	0.21	Dichlorvos	1 g L <sup>-1</sup>	100 mg L <sup>-1</sup>	5	50 mg g <sup>-1</sup>	Freundlich	0.99	Gheraout et al., 2020
3		18	Rhodamine B	2 g L <sup>-1</sup>	100–00 mg L <sup>-1</sup>	2.5–12	182.2 mg g <sup>-1</sup>	Langmuir	0.99	Deng et al., 2017
4	Gibbsite	44.1	Nortriptyline	100 m <sup>2</sup> L <sup>-1</sup>	0.1 mM	3–12	2.4 ± 0.1 μmol m <sup>-2</sup>	Langmuir	0.99	Sadri et al., 2018
5		5.82	Sb	10 g L <sup>-1</sup>	1 μmol L <sup>-1</sup>	5.5	38.66 mmol kg <sup>-1</sup>	Langmuir	0.98	Essington & Stewart, 2015
6		18	Se	0.8 g L <sup>-1</sup>	19 μmol L <sup>-1</sup>	3	9.5 mmol kg <sup>-1</sup>	Triple-layer model	-	Goldberg, 2014
7	Hematite	19.4	Phenol	5 g L <sup>-1</sup>	10 mg g <sup>-1</sup>	2–7	16.17 mg g <sup>-1</sup>	Langmuir	0.99	Dehghan and Anbia, 2017
8		100.5	Methylene blue and other metal ions - Cr, Co, Ni, Cu, Cd, Hg e Pb	0.62 g L <sup>-1</sup>	5–40 mg L <sup>-1</sup>	6.5	44.7 mg g <sup>-1</sup>	Langmuir	0.99	Sengupta et al., 2017
9		73.9	Congo Red	0.3 g L <sup>-1</sup>	400–100 mg L <sup>-1</sup>	-	628.9 mg g <sup>-1</sup>	Langmuir	0.99	Xu et al., 2012
10	Ilmenite	90.6	Methylene blue	0.03 g L <sup>-1</sup>	0.5–8 mg L <sup>-1</sup>	5.5	71.9 mg g <sup>-1</sup>	Langmuir	0.97	Chen, Jin, et al., 2011
11		152.3	Congo red, Methyl orange, and Methylene blue	0.3 g L <sup>-1</sup>	50–500 mg L <sup>-1</sup>	7.2	723.8, 150.7, and 54.5 mg g <sup>-1</sup>	Sips	0.98	Kang et al., 2018
12	Magnetite	44.82	Acid Red 18	0.75 g L <sup>-1</sup>	25–100 mg L <sup>-1</sup>	3	16.259 mg g <sup>-1</sup>	Freundlich	0.98	Berizi et al., 2016
13		23.35	Pb	10 mg L <sup>-1</sup>	110 mg L <sup>-1</sup>	6	41.66 mg g <sup>-1</sup>	Langmuir	0.99	Singhal et al., 2017
14		98.8	As	2.5 g L <sup>-1</sup>	45 mmol L <sup>-1</sup>	4.8	20.8 mg g <sup>-1</sup>	Langmuir		Daou et al., 2007
15		14.180	Tl	2 g L <sup>-1</sup>	20–100 mg L <sup>-1</sup>	6–9	1123 mg g <sup>-1</sup>	Temkin	0.98	Li and Arai, 2020

The removal of phosphate species from solution by adsorption onto natural-calcite was evaluated alongside the effect of pH, phosphate/mineral ratio, and contact time (Karageorgiou et al., 2007). The results indicated that pH plays an important role, and the adsorption process is more efficient in the basic pH region. The orthophosphate removal is almost complete for concentrations as high as 60 mg L<sup>-1</sup> of phosphate, which corresponds to 0.1 to 1 mM g<sup>-1</sup> pollutant to adsorbent ratio (Karageorgiou et al., 2007). Tannic acid, a calcite flotation agent widely used in mineral processing, had its mechanisms and behaviors studied on calcite surfaces, utilizing isothermal, kinetic, and thermodynamic studies. The adsorption capacity of tannic acid increased with initial concentration in the optimal pH 8. Sips isotherm model best predicted equilibrium data, while the adsorption kinetics followed a pseudo-second-order model, indicating that the adsorption process was controlled by chemical reaction. The maximum adsorption capacity was 13.55 mg g<sup>-1</sup>, considering 10 g L<sup>-1</sup> of calcite and 100 mg L<sup>-1</sup> of tannic acid (Karageorgiou et al., 2007). Moreover, other studies were conducted: regarding the dynamic of asphaltene adsorption on calcite packs (Karageorgiou et al., 2007); L-malic, D-malic, and succinic acid adsorption (Li et al., 2019a); determination of Ni isotope fractionation its adsorption processes (Castillo Alvarez et al., 2020); arsenate (Markovski et al., 2014a) and Cd(II) (Song et al., 2011) on modified calcite; as well as a molecular simulation study on the adsorption of hydrogen sulfide in calcite pores (Cai et al., 2020).

Even though calcite is the most commonly found mineral, dolomite is the most studied carbonate towards adsorption. The adsorption of several potentially toxic ions was tested on dolomite, such as those of the elements strontium and barium (Ghaemi et al., 2011), copper and lead (Pehlivan et al., 2009; Şenol and Şimşek, 2020), arsenate (Ayoub and Mehawej, 2007), chromium (Albadarin et al., 2012; Stefaniak et al.,

2000), cadmium and nickel (Mohammadi et al., 2015), and fluoride (Wongrueng et al., 2016). As an outcome, all studies warranted dolomite as a cheap and useful solid material, with good adsorption properties towards hazardous inorganic compounds (Stefaniak et al., 2000).

Siderite has been applied as a pristine mineral (Erdem and Özverdi, 2005; Hajji et al., 2019; Wang and Reardon, 2001), as well as the principal constituent in iron-rich sludge adsorbents (Qu et al., 2020; Sharma et al., 2013b; Zhu et al., 2018, 2019). By using stirred flow-through reactor experiments, siderite (90 m<sup>2</sup> g<sup>-1</sup>) adsorption of As(III), Cr(VI), and competitive As(III)/Cr(VI) was reinvestigated. As and Cr sorption isotherms fit the Langmuir model. Its competitive sorption responded to a sigmoidal Hill model. The maximum uptake for each pollutant was around 60 mg g<sup>-1</sup> (Hajji et al., 2019). Conversely, by submitting the wasted iron mud to hydrothermal treatment, a magnetic adsorbent removed other metals from smelting wastewater. The modification acted upon ferrihydrite's reductive dissolution to recombine the natural siderite, which was then re-oxidized to maghemite. As a result, at the dosage of 12.5 g L<sup>-1</sup>, a removal efficiency around 99% of Cu<sup>2+</sup>, Zn<sup>2+</sup>, Pb<sup>2+</sup>, and Cd<sup>2+</sup> was attained (Zhu et al., 2018).

Moreover, magnesite, smithsonite, aragonite, and bastnasite have all been applied to the wastewater treatment to some extent. Magnesite was employed in phosphate from municipal effluents (Masindi et al., 2016; Mavhungu et al., 2019) and as an adsorbent for cationic and anionic dyes (Ngulube et al., 2018). Lead (Zhang et al., 2019a), iron (Deng et al., 2017), and 8-hydroxyquinoline onto smithsonite have been studied (Cristina et al., 2011). As for aragonite, cadmium (Van et al., 2019; Yang et al., 2018), phosphate (Millero et al., 2001), and methanoic acid adsorption have been reported (Cooper and De Leeuw, 2002). Finally, alkyl hydroxamic acid's adsorption mechanism onto bastnasite was explored (Millero et al., 2001).

### 3. Clays

Clays are phyllosilicate minerals with layered structural units, constituted of one or two tetrahedral silica sheets around an octahedral aluminum sheet (Brigatti et al., 2006; Velde, 1995). They present small particle sizes, typically of less than 2  $\mu\text{m}$ , and high specific surface areas, as a result of their complex porous structures, which facilitates physical and chemical interactions with dissolved species (Bertagnolli et al., 2011; Djomgoue and Njopwouo, 2013; Luo and Daniel, 2003). Moreover, some specific factors determine how the adsorption process will occur onto clay minerals. Besides some of its specific properties, the contact time, clay dosage, and pH, play important roles (Otunola and Ololade, 2020).

Two essential topics on clay adsorption are its cation exchange capacity (Chapman, 2016) and specific surface area (Carter et al., 2018), because in this case, it is understood that they rule its adsorption capability (Dabrowski, 2001). Both properties are related: while cation exchange capacity translates as the total negative charge (Ammann et al., 2005; Lipson and Stotzky, 1983), the specific surface area is the total surface area with available adsorption sites (Kuila and Prasad, 2013; Macht et al., 2011). The larger the surface area, the greater is the cation exchange capacity (Ersahin et al., 2006; Kuila and Prasad, 2013). Besides, it is attractive to the point that variations found in these properties determinations can be attributed to the fact that some clay minerals have only external layers, such as kaolinite and illite, and others have internal and external layers, such as smectites and attapulgite (Hepper et al., 2006).

Another critical variable in adsorption studies is the contact time, which is how long the adsorbent needs to be in the same medium as the pollutant to achieve maximal removal efficiency (McKay, 1982; Suliestyah Hartami and Tuheteru, 2020; Yang et al., 2009). The adsorption rate of clays tends to increase with the increased contact time, remaining constant until equilibrium is reached (Yin and Zhu, 2016). Moreover, the contact time varies according to clay dosage (Otunola and Ololade, 2020). There are no specific guidelines determining clay dosage for wastewater treatment; therefore, previous studies and literature search is needed before application. It has been discussed that a small dosage, around 1 to 3  $\text{g L}^{-1}$ , of clay minerals, is enough to adsorb reasonable concentration pollutants from contaminated effluents (Kausar et al., 2018; Lazaratou et al., 2020).

Moreover, observations on the influence of the pH of the system demonstrated that, as pH increases, there is an immediate increase in the rate of adsorption (Akpomie and Dawodu, 2016; Chen et al., 2011b; Es-Sahbany et al., 2019; Otunola and Ololade, 2020; Potgieter et al., 2006). However, it must be emphasized that pH also regulates other processes, such as the chemical equilibrium of the target pollutant molecule with the clay (Beattie, 1956; Hackling and Garnett, 1985). Therefore, even though generally basic pH enhances the clays' adsorption capacity, this is not a rule (Györfi et al., 2020; Hamza et al., 2020; Maleki and Karimi-Jashni, 2020; Obayomi et al., 2020; Romdhane et al., 2020).

#### 3.1. Kaolinite

One of the most currently applied geological materials for adsorption belongs to the hydrous silicates class: kaolinite (Velde, 1995). Its general molecular formula is  $\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$ . Kaolinite is a layered silicate mineral, with one tetrahedral sheet of silica linked through oxygen atoms to one octahedral sheet of alumina octahedral (Essington and Stewart, 2015; Kuila and Prasad, 2013; Sadri et al., 2018).

Ghorbanzadeh et al. (2015) reported on the removal of arsenate and arsenite from aqueous solution. Kaolinite surface area was determined as  $10.1 \text{ m}^2 \text{ g}^{-1}$ . Adsorption isotherms were conducted with initial arsenic concentrations of 0.05, 0.06, 0.1, 0.15, and 0.2  $\text{mmol L}^{-1}$ , with 27  $\text{g L}^{-1}$  of the adsorbent. The equilibrium was studied for 24 h. Optimal adsorption pH for  $\text{As}^{5+}$  was found to be around 5 (99.9%), with the efficiency rates decreasing as pH increased. For  $\text{As}^{3+}$  adsorption, at pH 7,

the maximum adsorption capacity reached was  $380 \mu\text{g g}^{-1}$ . Both As(V) and As(III) adsorption isotherms fit the Langmuir model ( $R^2 = 0.968$  and  $0.979$ , respectively), and  $q_{\text{max}}$  values of  $0.203$  and  $0.241 \text{ mg g}^{-1}$  (Ghorbanzadeh et al., 2015). Mohapatra et al. (2007) observed similar responses while studying pH for As(V) adsorption onto kaolinite, and maximum adsorption was also achieved at pH 5.0. Equilibrium was reached within 3 h, and  $q_{\text{max}}$  value of  $0.86 \text{ mg g}^{-1}$  was found by fitting the Langmuir equation to the adsorption isotherms ( $R^2 = 0.98$ ). The authors also observed an adverse effect of an increase in temperature on As(V) adsorption; therefore, interactions are exothermic. The electrokinetic behavior of kaolinite was modified in the pollutant's presence, which can be concluded as inner-sphere surface complexation and strong specific ion adsorption (Mohapatra et al., 2007).

Chantawong et al. (2003) investigated the adsorption of  $\text{Cd}^{2+}$ ,  $\text{Cr}^{3+}$ ,  $\text{Cu}^{2+}$ ,  $\text{Ni}^{2+}$ ,  $\text{Pb}^{2+}$ , and  $\text{Zn}^{2+}$  by Thai kaolinite. Except for  $\text{Ni}^{2+}$ , adsorption increased as pH increased. Isotherms fit the Freundlich model, with  $R^2$  ranging from 0.93 to 0.99, for each ion. Adsorption followed the order:  $\text{Cr} > \text{Zn} > \text{Cu} \approx \text{Cd} \approx \text{Pb} > \text{Ni}$ , with maximum removal capacity of  $1.13 \times 10^{-3}$ ,  $1.07 \times 10^{-3}$ ,  $4.20 \times 10^{-4}$ ,  $6.22 \times 10^{-4}$ ,  $7.73 \times 10^{-5}$ , and  $5.11 \times 10^{-5} \text{ mmol g}^{-1}$ , respectively (Chantawong et al., 2003). Liu et al., 2016a focused on a wide range of individual Pb, Zn, and Cu concentrations, varying from 0.1 to 100 mM, at different pH conditions. Again, the best results were obtained in basic pH. Also, the isotherms fit the Freundlich and Langmuir models for kaolinite. The maximum removal capacity for Cu was  $1.82 \text{ mg g}^{-1}$ , for Zn was  $6.19 \text{ mg g}^{-1}$ , and Pb was  $14.47 \text{ mg g}^{-1}$  (Liu et al., 2015). Also, Coles and Yong (2002) conducted batch equilibrium tests on kaolinite suspensions adjusted to pH 4 and pH 6, aiming to remove  $\text{PbCl}_2$  and  $\text{CdCl}_2$ . Better results were obtained at pH 6. Interestingly, by studying the equilibrium curves of both metal retention versus suspension pH, combined with an analysis of the speciation of the metals, it was evidenced that both divalent ( $\text{Pb}^{2+}$  and  $\text{Cd}^{2+}$ ) and monovalent ( $\text{PbCl}^+$  and  $\text{CdCl}^+$ ) species were adsorbed (Coles and Yong, 2002).

Ghosh and Bhattacharyya (2002) employed raw kaolinite obtained from a local deposit focusing on methylene blue adsorption. The raw adsorbent showed considerable adsorption, superior to the calcinated version of the material. Experiments were conducted considering contact time of 3 h, pH 10, initial methylene blue concentration of  $15 \text{ mg L}^{-1}$ , and adsorbent dosage of  $0.8 \text{ g L}^{-1}$ . In this condition, raw kaolinite uptake reached  $13.99 \text{ mg g}^{-1}$ , and the isotherms fit well the Langmuir model (Ghosh and Bhattacharyya, 2002). Doğan et al. (2009) addressed Maxilon Yellow 4GL and Maxilon Red GRL dyes kinetic adsorption. The equilibrium was set after 150 min. The adsorption kinetics followed pseudo-second-order for both dyes, with  $k_2$  values up to  $107.87 \times 10^4 \text{ g mol}^{-1} \text{ min}^{-1}$  for Maxilon Yellow and  $72.09 \times 10^4 \text{ g mol}^{-1} \text{ min}^{-1}$  for Maxilon Red (Doğan et al., 2009). Despite kaolinite usual small surface area, several studies have been published on its efficiency towards dyes adsorption: aniline blue (Unuabonah et al., 2008a), basic fuchsin (Khan et al., 2015), crystal violet (Çiftçi et al., 2017; Dobrogowska et al., 1991; Khan et al., 2015; Sarma et al., 2019; Yariv et al., 1991), solophenyl yellow GFL (Kamel et al., 1991), solophenyl red 6BL (Kamel et al., 1991), diphenyl pink BF (Kamel et al., 1991), 9-aminoacridine (Harris et al., 2001, 2006a–c), 3,6-diaminoacridine (Harris et al., 2001, 2006a, 2006b, 2006c), azure A (Harris et al., 2001, 2006a, 2006b, 2006c), safranin O (Harris et al., 2001, 2006a, 2006b, 2006c), reactive blue 221 (Karaoğlu et al., 2010), basic red 5 (Alom et al., 2014), rhodamine B (Batista et al., 2014; Khan et al., 2017; Rosales et al., 2018), methylene blue (Ghosh and Bhattacharyya, 2002; Greathouse et al., 2015; Hang and Brindley, 1970; Mukherjee et al., 2015), malachite green (Castellini et al., 2008; Sarma et al., 2019), reactive red 120 (Abidi et al., 2017), congo red (Bhattacharyya et al., 2015; Shaban et al., 2018), and many others.

Moving on further than dyes, one study investigated their adsorption alongside to other 23 organic pollutants, such as acridine, 8-amino quinoline, quinoline, and pyridine (Harris et al., 2001). Organic pollutants are of great concern due to their ubiquitous occurrence,

even in remote ecosystems (Lu and Astruc, 2020; Sun et al., 2020; Zhang et al., 2003). Among them, pesticides are of great concern (Bache et al., 1964; Sharma et al., 2020). Clausen et al. (2001) investigated atrazine (6-chloro-N2-ethyl-N4-isopropyl-1,3,5-triazine-2,4-diamine), isoproturon ([3-(4-isopropyl phenyl)-(1,1-dimethylurea)]), mecoprop ((*RS*)-2-(4-chloro-2-methyl phenoxy)propionic acid), 2,4-D (2,4-dichloro-phenoxy-acetic acid), and bentazone (3-isopropyl-1H-2,1,3-benzothiadiazin-4-(3H)-one 2,2-dioxide) adsorption towards kaolin. Specific surface area and mineral surface charge proved important factors: detectable adsorption of anionic pesticides was only measured when positive sites were present on the mineral surface. When an electrolyte was added, detectable adsorption of mecoprop and 2,4-D was also measured on kaolinite, probably due to the formation of surface complexes. However, the adsorption of the uncharged pesticides, atrazine, and isoproturon, did not suffer interferences. Despite their efforts, pesticides adsorb weakly to pure kaolin in aqueous solutions (Clausen et al., 2001).

### 3.2. Montmorillonite

Montmorillonite is a clay mineral, whose molecular formula can be described as  $(\text{Na}, \text{Ca})_{0.33}(\text{Al}, \text{Mg})_2(\text{Si}_4\text{O}_{10})(\text{OH})_2(\text{H}_2\text{O})_n$ , with substantial isomorphous substitution. On behalf of adsorption, this mineral presents peculiar characteristics, such as fast cationic exchange and interlayer swelling (Bhattacharyya and Gupta, 2008). Cations such as  $\text{Na}^+$  and  $\text{Ca}^{2+}$  tend to form surface complexes; therefore, they are easily exchanged with solute ions (Dähn et al., 2002). In its structure, interlayer swelling occurs in aqueous media. This process depends on valences and the atomic radius of the exchangeable cations (Hennig et al., 2002), which leads to additional inner-sphere complexation and adsorption (Elzinga and Sparks, 1999). Although there is a conflict in the literature about whether bentonites are a sub-group of montmorillonites or simply the same thing, this review article will approach both subjects together. When it comes to writing reports on different knowledge areas, there is a deficiency sometimes in standardizing terms, which can result in less visibility for several relevant and well-performed studies, just because the authors know a specific material by a name other than the usual one, but that still is scientifically accepted (Malamis and Katsou, 2013).

Several studies report on montmorillonite adsorption towards metals (Álvarez-Ayuso and García-Sánchez, 2003; Andini et al., 2006; Angove et al., 1998; Bhattacharyya and Gupta, 2007a; Donat et al., 2005; Gupta et al., 2003; Gupta and Bhattacharyya, 2006; Jain and Sharma, 2002; Jobstmann and Singh, 2001; Kandah, 2004; Karapinar and Donat, 2009; Liu and Jiang, 2010; López et al., 1998; Mathialagan and Viraraghavan, 2002; Putro et al., 2017; Ulmanu et al., 2003; Wasewar et al., 2010). Gupta and Bhattacharyya (2006) reported the adsorption of  $\text{Cd}^{2+}$  onto raw and modified montmorillonite. The removal efficiency reached 94.5% for raw montmorillonite, compared to 64.8%, 81.0%, and 99.3% for ZrO/montmorillonite, tetrabutylammonium/montmorillonite and acid-activated montmorillonite, respectively. This removal efficiency can be translated as an adsorption capacity of  $32.7 \text{ mg g}^{-1}$ . Again, the adsorption of  $\text{Co}^{2+}$  was only slightly superior onto acid activated montmorillonite in comparison to the raw material, reaching 29.7 and  $28.6 \text{ mg g}^{-1}$  of adsorption capacity (Bhattacharyya and Gupta, 2007b). Considering the time, effort, and cost, modified montmorillonite is not offset by raw montmorillonite efficiency (Gupta and Bhattacharyya, 2006). In Table 4, several studies on montmorillonite adsorption towards metals are summarized.

The adsorption of several dyes onto both natural and modified montmorillonites has been studied over the years, such as methylene blue (Benhouria et al., 2015; Feddal et al., 2014; Hong et al., 2009; Kahr and Madsen, 1995; Wei et al., 2018; Zhang et al., 2019a), methyl violet (Fabryanty et al., 2017; Guiza et al., 2012; Puri and Sumana, 2018; Zhang et al., 2019b), methyl orange (Chen et al., 2011a; Leodopoulos et al., 2012; Zhang et al., 2019c), naphthol green (Zhang et al., 2019e), direct red 23 (Mahvi and Dalvand, 2020), diamond fast brown KE (El-Defrawy et al., 2019), reactive red 120 (Mahmoodi et al.,

2019; Tabak et al., 2010) and acid fuchsin (Cao et al., 2017; Gong et al., 2018). Data on dye adsorption can be summarized in results obtained by Fil et al. (2014), who studied Red Violet 3RN color removal in simulated wastewater using montmorillonite clay. They concluded that the process efficiency rate increased with increasing pH, temperature, dye concentration, and agitation speed but decreased with increased ionic strength and adsorbent dosage. Langmuir model better represented the equilibrium data. Also, kinetics fit the pseudo-second-order model (Fil et al., 2014).

Chang et al. (2015) investigated tetracycline adsorption, an antibiotic onto high-charge Ca-montmorillonite and low-charge Na-montmorillonite. The adsorption data obeyed Langmuir isotherm, with maximum pollutant adsorption of  $468$  and  $404 \text{ mg g}^{-1}$  on Ca and Na montmorillonites, respectively. The adsorption was endothermic, suggesting strong physical adsorption, which is expected for organic molecules adsorbents with high surface area and a small pore size ratio (Chang et al., 2015). Also, concerning natural montmorillonite, Thiebault and Boussafir (2019) explored the adsorption of the psychoactive drugs codeine, diazepam, and oxazepam onto Na-montmorillonite. At  $\text{pH} = 7.5$ , codeine cation exchange resulted in the highest amount adsorbed among the tests ( $q_{\text{max}} = 0.381 \text{ mmol g}^{-1}$ ). Therefore, their significant contribution was proving that variation in pH strongly impacts the pollutant and sorbent affinity (Thiebault and Boussafir, 2019). Also, studies report atenolol (Chang et al., 2019) and nortriptyline (Sadri et al., 2018) in raw Ca-montmorillonite, while 4-acetaminophenol (Chu et al., 2019), amoxicillin, and diclofenac (Khan et al., 2020) removal were reported for modified montmorillonite.

At last, pesticides adsorption onto montmorillonite was reported and verified as well, demonstrating that it acts as a natural scavenger of pesticides due to the abundant availability, large specific surface area, and high adsorptive and ion exchange properties (Bhardwaj et al., 2014; Cabrera et al., 2008; Kovacevic et al., 2011; Ozcan et al., 2012; Park et al., 2014; Suciú and Capri, 2009). Carbaryl, aldrin, malathion, 2,4-D, and dimethoate adsorption capacity onto raw montmorillonite were  $3.70$ ,  $14.96$ ,  $7.95 \text{ mg g}^{-1}$  and  $0.90$ ,  $0.96 \text{ } \mu\text{g kg}^{-1}$ , respectively (Al Kuisi, 2002; Chen et al., 2009; Ozcan et al., 2012; Pal and Vanjara, 2001; Park et al., 2019); 8-quinoline carboxylic acid adsorption capacity onto Na-, acidic- and organo-montmorillonite was  $65.4$ ,  $67.6$  and  $75.9 \text{ mg g}^{-1}$  (Mekhloufi et al., 2013); Na- and Ca-montmorillonite adsorption capacity towards phosmet reached  $1.04 \times 10^{-3}$  and  $1.648 \times 10^{-3} \text{ mg g}^{-1}$  (Sánchez Camazano and Sánchez Martín, 1983), while K-, Na- and Ca-montmorillonite towards phosdrin reached  $1.882 \times 10^{-3}$ ,  $3.548 \times 10^{-3}$  and  $2.150 \times 10^{-3} \text{ mg g}^{-1}$  (Sánchez Camazano and Sánchez Martín, 1983).

### 3.3. Illite

The term illite was proposed by (Grim et al., 1937) as a general term, not as a specific clay mineral name, to designate mica-like clay minerals of Illinois. Since then, the illite definition was progressively improved (Meunier and Velde, 2010). Thus, Illite is understood as a group of closely related non-expanding phyllosilicate. Its chemical formula is given as  $(\text{K}, \text{H}_3\text{O})(\text{Al}, \text{Mg}, \text{Fe})_2(\text{Si}, \text{Al})_4\text{O}_{10}[(\text{OH})_2, (\text{H}_2\text{O})]$ , with considerable ion substitution (Meunier et al., 2004; Meunier and Velde, 2010). In addition to the surface hydrogen bonds, illite cleavage releases potassium ions, which results in the surface of high-density ionic bonds. Thus, cleaved illite surfaces are moderately hydrophilic (Park et al., 2019; Zhang et al., 2018).

Picloram, a herbicide adsorbed on illite, was studied under pH variation. Batch tests were performed with an adsorbent dosage of  $16 \text{ g L}^{-1}$  and pollutant concentrations ranging from  $0.2$  to  $5.0 \text{ mmol L}^{-1}$ . The adsorption isotherms well-fit both Langmuir and Freundlich models, with  $q_{\text{max}}$  values of  $14 \pm 2$ ,  $12 \pm 3$ , and  $11 \pm 1 \text{ } \mu\text{mol g}^{-1}$  at pH 3, 5, and 7, respectively (Marco-Brown et al., 2019). The picloram molecule interactions with illite surfaces exhibited anionic profile adsorption, with a decrease in adsorption when the pH increases,

**Table 4**  
Different montmorillonites adsorption response on metal ions removal efficiency.

Type	Dosage (g L <sup>-1</sup> )	Pollutant – C <sub>0</sub>	q <sub>max</sub> (mg g <sup>-1</sup> )	Reference
Acid-activated	2	Cu <sup>2+</sup> 10–250 mg L <sup>-1</sup>	32.3	Bhattacharyya and Gupta, 2011
	2	Ni <sup>2+</sup> 10–250 mg L <sup>-1</sup>	29.5	Bhattacharyya & Gupta, 2008
Calcareous	1	Cu <sup>2+</sup> 10–100 mg L <sup>-1</sup>	12.97	Sdiri et al., 2012
Calcium	20	Cu <sup>2+</sup> 13–160 mg L <sup>-1</sup>	12.633	Wu et al., 2011
Illite system	3	Pb <sup>2+</sup> 100 mg L <sup>-1</sup>	54.12	Oubagaranadin et al., 2010
	17	Cu <sup>2+</sup> 10–150 mg L <sup>-1</sup>	30.99	Oubagaranadin and Murthy, 2010
K10	16	Ni <sup>2+</sup> 10–2000 mg L <sup>-1</sup>	2.10	Carvalho et al., 2008
Raw	2	Cu <sup>2+</sup> 10–250 mg L <sup>-1</sup>	31.8	Bhattacharyya and Gupta, 2011
	1	Cu <sup>2+</sup> 10–100 mg L <sup>-1</sup>	17.89	Sdiri et al., 2011
	2	Fe <sup>3+</sup> 10–250 mg L <sup>-1</sup>	28.9	Bhattacharyya and Gupta, 2008
	2.5	Pb <sup>2+</sup> 150 mg L <sup>-1</sup>	57.0	Zhang and Hou, 2008
	2	Ni <sup>2+</sup> 10–250 mg L <sup>-1</sup>	28.4	Bhattacharyya and Gupta, 2008
	1	Fe <sup>3+</sup> , Co <sup>2+</sup> , Ni <sup>2+</sup> 50 mg L <sup>-1</sup>	22.6	Bhattacharyya and Sen Gupta, 2009
Tetrabutylammonium	2	Fe <sup>3+</sup> 10–250 mg L <sup>-1</sup>	19.7	Bhattacharyya and Gupta, 2008b
ZrO	2	Fe <sup>3+</sup> , Co <sup>2+</sup> , Ni <sup>2+</sup> 50 mg L <sup>-1</sup>	23.8	Bhattacharyya and Gupta, 2008c

which is very interesting, considering that the illite point of zero-charge is around 2.8 (Marco-Brown et al., 2019; Park et al., 2019). Tetracycline is a semi-synthetic antibiotic commonly detected in soils due to animal-produced slurry applications as fertilizer (Agersø et al., 2006). Chang et al. (2012) reported illite adsorption capacity towards tetracycline adsorption at 32 mg g<sup>-1</sup>, at pH 5–6. Adsorption equilibrium data obeyed the Freundlich isotherm model. Therefore, considering the q<sub>max</sub> value reported in this study, illite may act as an essential environmental deposit on fate, routes, and transportation of antibiotics in soils (Chang et al., 2012). Although the studies discussed above, as well as the study carried out by (Fil et al., 2016) regarding methyl violet dye, point to non-linear adsorption models, it has also been reported that for larger molecules, such as microcystin-LR, the application of linear isotherms might be proven more suitable (Liu et al., 2019).

Ionic adsorption onto illite has been inspected as well. For example, uranium sorption characteristics tend to be dominated by chemical ion-exchange, an endothermic and spontaneous process that increases entropy (Liao et al., 2020). Fernandes and Baeyens (2019) modeled lead adsorption considering two sites protolysis non-electrostatic surface complexation and cation exchange, and reported maximal uptake values 4.0 × 10<sup>-2</sup> to 2.0 × 10<sup>-3</sup> mol kg<sup>-1</sup> (Fernandes and Baeyens, 2019). Moreover, investigations on copper and nickel adsorption on illite/smectite systems were reported by (Du et al., 1997), Rosskopfová's et al. (2016), and Viglašová et al. (2017). In addition to illite ionic adsorption, thallium and strontium adsorption have been investigated as well, but will not be explored in this review for our scope is environmental remediation, and such metals are not yet considered environmental relevant contaminants (Galamboš et al., 2013; Wick et al., 2018; Zhang et al., 2016).

#### 4. Zeolites

Zeolites of natural origin are formed due to volcanic activity, either by water or deposition of volcanic dust in saline deposits (Delkash et al., 2015; Malamis and Katsou, 2013). They are complex, microporous

inorganic polymers formed by an infinitely extensive three-dimensional structure of silicon and aluminum tetrahedrons linked together by sharing oxygen (Hong et al., 2019; Niu et al., 2020). The most commonly found natural types are mordenite, chabazite, and clinoptilolite (Dehghan and Anbia, 2017; Flanigen, 1991). The zeolite crystal structure's inner cavities are generally in the range of 0.5–1.2 nm, a factor determined by formation route and different Si/Al ratios. This Si/Al ratio acts directly on the thermal resistance of zeolites and cation exchange properties: low-silica zeolites with Si/Al ratio below 2 have excellent ion exchange capacity; however, the higher the aluminum content, the less stable tends to be the zeolite (Jiang et al., 2018; Wen et al., 2018). Unlike geopolymers, zeolites, despite having the same constitution, are highly crystalline materials with higher thermal and chemical resistance. Another difference is that zeolites need a higher amount of water present in their formation reaction medium. The synthesis reactions take longer than that of the geopolymers, much due to the crystallization stage (Fernández-Jiménez et al., 2005).

Jafari-zare and Habibi-yangjeh (2010) studied the adsorption of rhodamine B and methylene blue in both single and binary component systems, using natural zeolite formed predominantly by clinoptilolite. The experiments were carried out at pH 12, with an adsorbent dosage of 0.2 g L<sup>-1</sup> and initial dye concentration of 6 × 10<sup>-6</sup> mol L<sup>-1</sup>. Results showed that the maximum capacity of clinoptilolite for the dyes methylene blue and rhodamine B in a single system was 7.95 × 10<sup>-5</sup> and 1.26 × 10<sup>-5</sup>, respectively, with both isotherms being better described by the Langmuir model, with determination coefficients (R<sup>2</sup>) greater than 0.991. A reduction in the adsorption capacity of both dyes was verified in the binary system due to the competition of the molecules for the active sites available in the adsorbent; thus, the q<sub>max</sub> value for rhodamine B was 2.2 × 10<sup>-6</sup> mol g<sup>-1</sup>, while for methylene blue was 7.44 × 10<sup>-5</sup> mol g<sup>-1</sup>, fitted again by Langmuir model (R<sup>2</sup> of 0.984 for the rhodamine B and 0.997 for the methylene blue) (Jafari-zare and Habibi-yangjeh, 2010).

Natural zeolites can also be used as adsorbents to remove pharmaceuticals in an aqueous solution. de Sousa et al. (2018) removed azithromycin, ofloxacin, and sulfamethoxazole using faujasite zeolites

with different Si/Al ratios. The zeolite FAU-1 had a Si/Al ratio of 30 and a surface area of  $914 \text{ m}^2 \text{ g}^{-1}$ , while the FAU-2 had a Si/Al ratio of 82 and a surface area of  $899 \text{ m}^2 \text{ g}^{-1}$ . The adsorption tests were carried out with initial concentrations of antibiotics ranging from 10 to  $400 \mu\text{g L}^{-1}$ , for 360 min, with a solution volume of 30 mL. The results showed that the FAU-1 zeolite, with greater surface area, showed greater adsorption capacity for both antibiotics, within the range of  $8.5 \text{ mg g}^{-1}$  for azithromycin to  $31.32 \text{ mg g}^{-1}$  for ofloxacin. A study simulating an effluent containing the three antibiotics, under conditions of pH 7.25, an adsorbent dosage of  $10 \text{ mg L}^{-1}$ , a contact time of 2 h, initial concentration of azithromycin, ofloxacin, and sulfamethoxazole of 391, 378, and  $78.6 \text{ ng L}^{-1}$ , respectively, showed that zeolite FAU-1 responds well against the treatment of effluents contaminated with pharmaceuticals, removing up to 60% of azithromycin, 76% ofloxacin and 44% of sulfamethoxazole (de Sousa et al., 2018).

Due to zeolite's tetrahedral aluminum structure, a negative surface charge is created in the mineral network structure. The presence of cations can counterbalance this charge, usually,  $\text{Ca}^{2+}$ ,  $\text{Na}^+$  and  $\text{K}^+$ , located in the cavities; therefore, these cations are exchangeable with other cations, including environmental pollutant metals ions (Malamis and Katsou, 2013; Sprynskyy et al., 2006). For further understanding, the works on  $\text{Ag}^+$ ,  $\text{Cd}^{2+}$ ,  $\text{Co}^{2+}$ ,  $\text{Cr}^{3+}$ ,  $\text{Cr}^{6+}$ ,  $\text{Cu}^{2+}$ ,  $\text{Fe}^{3+}$ ,  $\text{Mn}^{2+}$ ,  $\text{Ni}^{2+}$ ,  $\text{Pb}^{2+}$ , and  $\text{Zn}^{2+}$  adsorption are summarized in Table 5, displaying the most important topics and specifications of each research (Akgül et al., 2006; Álvarez-Ayuso et al., 2003; Álvarez-Ayuso and García-Sánchez, 2003; Baker et al., 2009; Bhattacharyya and Gupta, 2007a; Erdem et al., 2004; Ijagbemi et al., 2009; Malamis and Katsou, 2013; Minceva et al., 2008; Motsi et al., 2009; Ören and Kaya, 2006; Ren et al., 2016a; Sari and Tüzen, 2013; Sen Gupta and Bhattacharyya, 2008; Sheta et al., 2003; Sprynskyy et al., 2006; Yakout and Borai, 2014).

## 5. Geopolymers

Geopolymers are amorphous or semi-crystalline materials formed from polymerization reactions between geological originated materials containing aluminum and silicon, such as clays and fly ash, and an activating agent, usually strong bases as sodium or potassium hydroxide (Alouani et al., 2018; Bagci et al., 2017). Initially, geopolymers were

used as additives to Portland cement because they improved their mechanical properties and durability since geopolymers are highly resistant to acid attacks and are resistant to frost/defrost cycles and fire-resistant (Atiş et al., 2015; Komnitsas, 2011).

The large  $\text{OH}^-$  ion concentration in the reaction medium of geopolymer preparation is responsible for the amorphous three-dimensional network's dissolution, commonly found in the starting materials (Khale and Chaudhary, 2007). In more detail, the activating agent breakdown the solid aluminosilicate by repulsion caused by the strong base cations ( $\text{Na}^+$  or  $\text{K}^+$ ). In monomers, Al is tetrahedrally coordinated, and polycondensation reactions follow, forming a network composed of  $\text{SiO}_4$  and  $\text{AlO}_4$  tetrahedrons linked together by oxygen atoms. In this network, the alkali agent cation acts as a charge compensator in the geopolymer structure, enhancing adsorption possibilities (Duxson et al., 2007; Rahier et al., 2007). Studies involving the use of geopolymers as adsorbents for environmental remediation purposes, despite recent, are widely disseminated in literature, as presented in Table 6, and mainly focus on the removal of dyes molecules of aqueous solutions (Acisli et al., 2020; Alouani et al., 2018; Barbosa et al., 2018; Hua et al., 2020; Li et al., 2006; Rossatto et al., 2020; Schadeck Netto et al., 2019; Siyal et al., 2018) and towards metal ions (Al-Zboon et al., 2011; Chen et al., 2019; Cheng et al., 2012a; Kara et al., 2017; Liu et al., 2016c; Naghsh and Shams, 2017).

Barbosa et al. (2018) prepared a geopolymer using calcined kaolinite and rice husk ash as precursor materials and tested it versus methyl violet dye adsorption from aqueous solutions. The  $62 \text{ m}^2 \text{ g}^{-1}$  surface area as-synthesized geopolymer showed a  $q_{\text{max}}$  capacity of  $277 \text{ mg g}^{-1}$ , at pH 4.5, at a temperature of 328 K, and an adsorbent dosage of  $1.5 \text{ g L}^{-1}$ . Isotherms were better described by the Sips model, with  $R^2$  of 0.995, 0.989, 0.993, and 0.996 for the respective temperatures of 298, 308, 318, and 328 K (Barbosa et al., 2018). The endothermic character, which guarantees an improvement in the adsorption capacity by greater exposure of the geopolymer's active sites, was also reported by Alouani et al. (2018). They synthesized a geopolymer from fly ash from a thermoelectric plant for the adsorption of crystal violet dye. After 120 min, pH 5, and adsorbent dosage of  $1 \text{ g L}^{-1}$ , the maximum adsorption capacity of  $37 \text{ mg g}^{-1}$  was found, with the Langmuir model best describing the adsorption isotherms, with  $R^2 = 0.999$ , much higher

**Table 5**  
Specifications of studies on  $\text{Ag}^+$ ,  $\text{Cd}^{2+}$ ,  $\text{Co}^{2+}$ ,  $\text{Cr}^{3+}$ ,  $\text{Cr}^{6+}$ ,  $\text{Cu}^{2+}$ ,  $\text{Fe}^{3+}$ ,  $\text{Mn}^{2+}$ ,  $\text{Ni}^{2+}$ ,  $\text{Pb}^{2+}$ , and  $\text{Zn}^{2+}$  adsorption by different zeolites.

Material	Surface area ( $\text{m}^2 \text{ g}^{-1}$ )	Metal	Adsorbent dosage ( $\text{g L}^{-1}$ )	Initial concentration ( $\text{mg L}^{-1}$ )	pH	Temperature (K)	Adsorption capacity ( $\text{mg g}^{-1}$ )	Reference
Chabazite	1100	$\text{Zn}^{2+}$	20		6.0	293	7.98	Sheta et al. (2003)
	73.61	$\text{Cd}^{2+}$	2	50	5.5	298	120	Yakout and Borai (2014)
	–	$\text{Co}^{2+}$	5	30	5.0	298	1.50	Ouki and Kavannagh (1997)
		$\text{Cu}^{2+}$					3.80	
	17.83	$\text{Pb}^{2+}$	10	500	6.0	318	22.88	Minceva et al. (2008)
		$\text{Zn}^{2+}$					4.17	
		$\text{Cd}^{2+}$					5.46	
	–	$\text{Mn}^{2+}$	20	400	7.0	303	4.22	Erdem et al. (2004)
	20.3	$\text{Cd}^{2+}$	10	200	6.0	295	4.60	Álvarez-Ayuso et al. (2003)
		$\text{Cu}^{2+}$			5.0		5.91	
	$\text{Cr}^{3+}$			4.0		4.12		
	$\text{Fe}^{3+}$	37	400	3.5	295	6.61	Motsi et al. (2009)	
	$\text{Cu}^{2+}$			3.5		3.37		
	$\text{Ag}^+$	2	150	4.0	298	33.23	Akgül et al. (2006)	
Clinoptilolite/quartz	15.78	$\text{Pb}^{2+}$	12	80	4.0	308	10.00	Ren et al. (2016a)
		$\text{Cr(VI)}$	25				0.34	
		$\text{Zn}^{2+}$	1	20	4.0	298	6.00	Ören and Kaya (2006)
	$\text{Cd}^{2+}$			5.5		30.7		
	$\text{Ni}^{2+}$			5.7		21.1		
Phillipsite	961	$\text{Zn}^{2+}$	20	500	6.0	293	1.70	Sheta et al. (2003)
Vermiculite	14.6	$\text{Ni}^{2+}$	2.5	100	6.0	295	19.3	Álvarez-Ayuso and García-Sánchez (2003)
	0.84	$\text{Ag}^+$	20	400	4.0	293	46.2	Sari and Tüzen (2013)
Zeolite	–	$\text{Zn}^{2+}$	5	20	6.0	300	62.9	Baker et al. (2009)
	–	$\text{Pb}^{2+}$					56.82	



**Table 6**  
Summarization of recent studies involving geopolymers as adsorbents for environmental remediation purposes.

Adsorbent	Adsorbate	Surface area ( $\text{m}^2 \text{g}^{-1}$ )	$q_{\text{max}}$ ( $\text{mg g}^{-1}$ )	Reference
Fly ash	Methylene blue	–	37.04	Alouani et al., 2018
	$\text{Pb}^{2+}$	20.48	118.6	Liu, Huang, et al., 2016
	$\text{Cd}^{2+}$	130.45	26.246	Javadian et al., 2015
	$\text{Cs}^+$	114.16	15.24	Lee et al., 2016
Linz Donawitz converted slag	$\text{Ni}^{2+}$	30.84	85.29	
	Methyl violet	62	276.9	Barbosa et al., 2018
Mesoporous Metakaolin	Methyl Orange	35.66	0.339	Fumba et al., 2014
	$\text{NH}_4^+$	22.4	21.07	Luukkonen et al., 2016
	$\text{NH}_4^+$	19.3	19.7	
	$\text{Zn}^{2+}$	39.24	74.53	Kara et al., 2017
	$\text{Ni}^{2+}$	39.24	42.61	Kara et al., 2017
	$\text{Cd}^{2+}$	65.7	75.74	Cheng et al., 2012
	$\text{Co}^{2+}$	39.24	69.23	
	$\text{Mn}^{2+}$	39.24	72.34	
	$\text{Cu}^{2+}$	53.95	52.63	
	$\text{Pb}^{2+}$	100.99	629.21	
Mn-CuO/Graphene bottom ash	Direct sky blue	18.45	0.497	
Modified metakaolin	$\text{Cu}^{2+}$	216	40	Singhal et al., 2017

if compared to the fit of Freundlich ( $R^2 = 0.694$ ), Temkin ( $R^2 = 0.866$ ) and Dubinin-Radushkevich ( $R^2 = 0.555$ ) models (Alouani et al., 2018).

The addition and impregnation of magnetic agents, such as magnetite or zero-valent iron, was presented as a viable and innovative alternative to facilitate the separation of the adsorbent from the aqueous medium (Schadeck Netto et al., 2019; Hua et al., 2020); Rossatto et al. (2020) synthesized a magnetic geopolymer using silica, calcined kaolinite, sodium hydroxide, and magnetite to adsorb acid green 16 dye. The  $53 \text{ m}^2 \text{g}^{-1}$  surface area magnetic geopolymer presented a maximum adsorption capacity of  $400 \text{ mg g}^{-1}$  of dye at 328 K, at pH 2.3, and an adsorbent dosage of  $0.75 \text{ g L}^{-1}$ , with best adsorption isotherms described by the BET model with values of  $R^2$  from 0.96 to 298 K and 0.97 to 328 K (Rossatto et al., 2020).

El-eswed et al. (2012) studied the adsorption of  $\text{Cu}^{2+}$ ,  $\text{Ni}^{2+}$ ,  $\text{Zn}^{2+}$ ,  $\text{Cd}^{2+}$ , and  $\text{Pb}^{2+}$  ions in kaolinite/zeolite-based-geopolymers; under pH 4, adsorbent dosage of  $1 \text{ g L}^{-1}$ , 298 K, and contact time of 24 h. It was concluded that the adsorption is preferred against small metal ions, such as  $\text{Cu}^{2+}$ ,  $\text{Ni}^{2+}$ ,  $\text{Zn}^{2+}$ , concerning the larger ones, such as  $\text{Cd}^{2+}$  and  $\text{Pb}^{2+}$ . The values of the maximum adsorption capacities found by the Langmuir model's application varied from  $0.6 \text{ mmol g}^{-1}$  for  $\text{Cu}^{2+}$  to  $0.1 \text{ mmol g}^{-1}$  of  $\text{Cd}^{2+}$ . Besides, in ionic competition systems, the adsorption of  $\text{Cu}^{2+}$  and  $\text{Pb}^{2+}$  in geopolymers did not decrease, indicating cooperative adsorption, which is very interesting from possible interferences in the adsorption of the targeted pollutants (El-eswed et al., 2012).

## 6. Modified materials and composites

This final section presents an overview of the most relevant topics on modified geological originated materials. This straightforward approach was chosen to illuminate future perspectives while still focusing on natural and raw forms of application. Nevertheless, there are many ways of modifying geological originated materials aspiring to increase in adsorption capacity compared to the raw mineral.

Although in several studies previously discussed, the  $q_{\text{max}}$  values reported were adequate for untreated materials, many other comparative studies have shown that modifications, such as metal impregnation, or treatments, such as thermal or acid-base activation, can critically enhance adsorption capacity (Bhattacharyya et al., 2015; Bhattacharyya and Gupta, 2008; Bhattacharyya and Sen Gupta, 2009; Carvalho et al., 2008; Kausar et al., 2018; Martin et al., 2019; Obayomi et al., 2020; Park et al., 2014; Sen Gupta and Bhattacharyya, 2014; Singhal et al.,

2017; Suci and Capri, 2009; Unuabonah et al., 2008a,b; Wu et al., 2011). Even simple pre-treatments can extract soluble organic compounds and enhance chelating efficiency (Gaballah et al., 1997). Modifications methods comprise various kinds of modifying agents: bases, such as sodium hydroxide, calcium hydroxide, and sodium carbonate; mineral and organic acid solutions, such as hydrochloric acid, nitric acid, sulfuric acid, tartaric acid, citric acid, and thioglycolic acid, organic compounds, such as ethylenediamine, formaldehyde, epichlorohydrin, and methanol, and even oxidizing agents, such as hydrogen peroxide (Wan Ngah and Hanafiah, 2008).

### 6.1. Metal ions

Aiming U(VI) adsorption, Li et al. (2014) prepared thermally activated sodium by calcinating micron Na-feldspar at  $450 \text{ }^\circ\text{C}$  for 45 min, which resulted in a material with a larger specific surface area and larger porosity than the original. The prepared material presented maximum adsorption efficiency of  $0.5 \text{ mg L}^{-1}$  of  $\text{U}^{6+}$ , the equivalent to 95.49%, at pH 5.0, 318 K, and contact time of 600 min. The relationship between  $q_{\text{max}}$  and equilibrium concentration,  $C_e$ , was well described by the Freundlich model (Li et al., 2014). Ahmad and Mirza (2017) synthesized a novel ecofriendly alginate/Au/mica bio nanocomposite with remarkable environmental remediation capacity of  $\text{Pb}^{2+}$  and  $\text{Cu}^{2+}$  in a single and binary system. The bionanocomposite adsorption experiments data best fit the Freundlich isotherm model for  $\text{Pb}^{2+}$ , whereas for  $\text{Cu}^{2+}$  Langmuir isotherm model was more suitable, both single and binary systems, with a maximum adsorption capacity of 224.97 and  $169.817 \text{ mg g}^{-1}$ , respectively. Adsorption process was found to be endothermic and spontaneous, thus, the nanocomposite can be applied successfully to industrial wastewaters.

Hematite and ilmenite modified composites have been studied and proposed by several authors (Chen et al., 2017; Kang et al., 2018; Lee et al., 2013a; Ma et al., 2018; Sengupta et al., 2017; Shu et al., 2019; Waanders et al., 2016; Wang et al., 2019a; Xu et al., 2017, 2018; Yousef, 2017). Sengupta et al. (2017) prepared a mesoporous composite consisting of hematite micro sheets modified and decorated with tetragonal zirconia nanocrystallites, useful for adsorbing cationic pollutants from water, by a simple hydrothermal method. It exhibited improved adsorption behavior of various metal ions, with about 99% removal efficiency reached for  $\text{Cu}^{2+}$ ,  $\text{Cd}^{2+}$ , and  $\text{Pb}^{2+}$  at an initial concentration ranging from 5 to  $20 \text{ mg L}^{-1}$ , in comparison to unmodified zirconia and hematite. Wang et al., 2019b synthesized using ilmenite

and NaOH as starting materials a nanosized titanate composite by the solid-state route. After calcination at 900 °C, the final molecular formula of the composite was reached:  $\text{Na}_{0.79-0.82}\text{Fe}_{0.96-0.98}\text{Mg}_{0.07-0.08}\text{TiO}_{3.85-3.88}$ . The adsorption of  $\text{Cu}^{2+}$ ,  $\text{Cd}^{2+}$ , and  $\text{Pb}^{2+}$  ions fit pseudo-second-order adsorption kinetics and the Langmuir adsorption model. At 20 °C, the maximum adsorption capacities of the as-synthesized nanocomposite for  $\text{Cu}^{2+}$ ,  $\text{Cd}^{2+}$ , and  $\text{Pb}^{2+}$  were 13.8, 8.8, and 20.3  $\text{mg g}^{-1}$ , respectively. The thermodynamic parameters also revealed that these ions' adsorption was spontaneous and endothermic (Wang et al., 2019b). Interestingly, on the contrary to hematite and ilmenite, magnetite is usually used to cover other adsorbents to increase their magnetization capacity and adsorption (Amara et al., 2017; Chiou et al., 2017; Çiftçi et al., 2017; Malik et al., 2020; Tahergerabi et al., 2016; Talebzadeh et al., 2017). It has also been applied as the lead material for adsorption purposes, as by Mahdavian and Mirrahimi (2010). They proposed anchoring polyacrylic acid on superparamagnetic magnetite nanoparticles through surface modification for efficient separation of heavy metal cations from aqueous solutions. Four cations were studied:  $\text{Cu}^{2+}$ ,  $\text{Pb}^{2+}$ ,  $\text{Ni}^{2+}$ , and  $\text{Cd}^{2+}$ . For the prepared cation solutions at the constant thermal, time, and pH conditions, the maximum adsorption capacity was found to be for  $\text{Pb}^{2+}$  ( $40 \pm 0.55 \text{ mg g}^{-1}$ ), and the minimum was for  $\text{Cd}^{2+}$  ( $7 \pm 0.6 \text{ mg g}^{-1}$ ) (Mahdavian and Mirrahimi, 2010).

Adsorption of As(V) onto activated siderite was reported by Zhao and Guo (2014), with  $q_{\text{max}}$  values reaching 2.19  $\text{mg g}^{-1}$  estimated from Langmuir isotherm at 25 °C (Zhao and Guo, 2014). Calcite modifications resulted in novel porous bentonite and calcite-biochar composite, used for lead adsorption, among other studies (Markovski et al., 2014b; Mohammadi and Sedighi, 2013; Ramola et al., 2020). Under optimized conditions, 0.07 g of the adsorbent composite effectively removed 97.06% of 232  $\text{mg L}^{-1}$  after 3.5 h at pH of 5.5, and the maximum adsorption capacity reported was 500  $\text{mg g}^{-1}$  (Ramola et al., 2020). Suzuki et al. (2004) investigated the construction of a uniform three-dimensional network porous structure of  $\text{CaZrO}_3/\text{MgO}$  composites, synthesized using reactive sintering of highly pure mixtures of natural dolomite ( $\text{CaMg}(\text{CO}_3)_2$ ) and synthesized zirconia powders, with  $\text{LiF}_{(\text{aq})}$  additive. The pore-size distribution was set at around 1  $\mu\text{m}$ , and the porosity was controllable (about 30% to 50%) through different sintering temperatures (Suzuki et al., 2004). Porosity is a critical factor, and Stefaniak et al. (2002) reported as well on the influence of preparation conditions on adsorption properties and porosity of dolomite-based sorbents. The thermal decomposition of carbonates has a dominant role in creating a new solid phase: as temperature increases over 800 °C, the porosity decreases (Stefaniak et al., 2002). This controlled tailoring form has been investigated towards phosphate adsorption on Al/dolomite/montmorillonite composite (Gao et al., 2013) and dolomite-alginate composite beads (Huang et al., 2019), towards  $\text{Cr}^{6+}$  onto thermally treated dolomite and  $\text{Pb}^{2+}$  onto chitosan-dolomite composite beads (Şenol and Şimşek, 2020).

Without a doubt, modified clays are the most explored geological materials, according to how much data on the subject has been published compared to previously presented minerals (Uddin, 2017). The modified forms of kaolinite provided excellent results in reported studies. The adsorption of  $\text{Pb}^{2+}$  onto tripolyphosphate-modified kaolinite clay (Unuabonah and Adebawale, 2009) and 25% (w/w) aluminum sulfate modified kaolinite clay (Jiang et al., 2009) were investigated. In the first study, the mechanism suggested that adsorption possibly took place at the negatively charged sites. Thus, the presence of Penta sodium tripolyphosphate adsorbed onto the kaolinite clay may contribute to enhancing the adsorption capacity of the proposed adsorbent (Unuabonah and Adebawale, 2009). In the second report, the maximum adsorption capacity of  $\text{Pb}^{2+}$  adsorption onto the modified kaolinite was 20  $\text{mg g}^{-1}$ , about 4.5 times higher than the amount adsorbed onto unmodified kaolin, of 4.2  $\text{mg g}^{-1}$ , under the optimized conditions (Jiang et al., 2009). Moreover, aiming to remove  $\text{Pb}^{2+}$ ,  $\text{Zn}^{2+}$ , and  $\text{Cd}^{2+}$ , sodium polyphosphate (SPP) modified kaolinite clay was synthesized and evaluated, indicating that increase in temperature was the dominant

parameter for increasing adsorption capacity (Amer et al., 2010). Nano-magnetite/kaolinite composite with kaolinite were produced and tested against Cu, Pb, Cd, Cr and Ni ions adsorption. The magnetic composite capacity increased over time and over increased pH (Lasheen et al., 2016).

Modified forms of montmorillonite clays have been studied and applied to the adsorption of Cu, Pb, Hg, Cd, Cs, Zn, Fe, Co, Ni, Sr, As, Cr, Fe, and even Pu (Abou-El-Sherbini and Hassanien, 2010; Begg et al., 2013; Bhattacharyya and Sen Gupta, 2009; Cruz-Guzmán et al., 2006; Goncharuk et al., 2010; Karamanis and Assimakopoulos, 2007; Liu et al., 2016a; Ma et al., 2015; Na et al., 2010; Oubagaranadin et al., 2010; Park et al., 2012; Sen Gupta and Bhattacharyya, 2009; Soltermann et al., 2013; Vinuth et al., 2015; Xu et al., 2008; Yu et al., 2008, 2009; Pereira et al., 2013). Organically modified montmorillonite clay was used to remove  $\text{Cu}^{2+}$ ,  $q_{\text{max}} = 62.5 \mu\text{g g}^{-1}$  (Abou-El-Sherbini and Hassanien, 2010). Montmorillonites modified with natural organic cations were synthesized and tested for the adsorption of  $\text{Pb}^{2+}$  and  $\text{Hg}^{2+}$  by Cruz-Guzmán et al. (2006). Carbon modified aluminum-pillared montmorillonite has shown good uptake of  $\text{Cd}^{2+}$  from an aqueous system,  $q_{\text{max}} = 161.75 \text{ g g}^{-1}$  (Yu et al., 2008). Al-13 pillared montmorillonites were also prepared with different Al to clay ratios to remove  $\text{Cd}^{2+}$  and  $\text{PO}_4^{3-}$  from aqueous solution (Ma et al., 2015). Aluminum-pillared-layered montmorillonites also proved their potential as a sorbent in removing  $\text{Cu}^{2+}$  and  $\text{Cs}^+$  from aqueous solutions (Karamanis and Assimakopoulos, 2007). Acid-activated montmorillonite-illite was examined for removing  $\text{Cu}^{2+}$  and  $\text{Zn}^{2+}$  from industrial wastewater containing  $\text{Pb}^{2+}$  at low concentration (Oubagaranadin et al., 2010). Calcined tetrabutylammonium bromide modified montmorillonite was prepared and used for the adsorption of  $\text{Fe}^{3+}$ ,  $\text{Co}^{2+}$ , and  $\text{Ni}^{2+}$  from aqueous solution (Bhattacharyya and Sen Gupta, 2009).

The reports on montmorillonites modifications also include thermal (Aytas et al., 2009; Zuo et al., 2017), acid/base chemical treatment (Akpomie and Dawodu, 2016; Shawabkeh et al., 2007), polycyclic-aromatic hydrocarbon (Biswas et al., 2016), phosphate (Park et al., 2012), polyethyleneimine (Goncharuk et al., 2010), Ti-pillared (Na et al., 2010), chitosan-beads (Arvand and Pakseresht, 2013; Liu et al., 2015; Pereira et al., 2013), cysteine (El Adraa et al., 2017; Stathi et al., 2007), goethite (Olu-Owolabi et al., 2010), alkyl benzene sulfonate (Sandy Maramis et al., 2012), 3-aminopropyltriethoxysilane (Guerra et al., 2013) and 3,2-aminoethylamino-propyl-trimethoxysilane (Guerra et al., 2013), manganese oxide (Eren and Afsin, 2008), amine and carboxylate (Anirudhan et al., 2012), aluminum pillaring (Arfaoui et al., 2008), thermal-hydro-mechanical (Lee et al., 2013b), magnesium ferrite nanoparticles (Kaur et al., 2015) and humic acid (Wu et al., 2011).

Zeolites modifications can be separated into two larger groups: organic and inorganic treatments. For the first group, reports include hexadecyltrimethylammonium bromide-modified NaY zeolite for both cationic and oxyanionic metal ions (Chao and Chen, 2012), amine-modified zeolite for Pb and Cd ions adsorption (Wingenfelder et al., 2005), surfactant-modified natural zeolites for  $\text{As}^{5+}$  adsorption (Chutia et al., 2009), thiourea modified synthetic zeolite X and its adsorption of  $\text{Cd}^{2+}$  (Zhang et al., 2019a), cetylpyridinium bromide modified zeolite for Hg ion adsorption (Liu et al., 2016b), ethylenediamine and monoethanolamine modified synthesized  $\beta$ -zeolite for adsorption of  $\text{Pb}^{2+}$  (Motlagh Bahadory Esfahani and Faghihian, 2014), hexadecylpyridinium bromide modified zeolite for  $\text{Cr}^{6+}$  adsorption (Zeng et al., 2010), surfactant modified zeolite for the selective adsorption of  $\text{Pb}^{2+}$  and  $\text{Cr}^{6+}$  (Ren et al., 2016b) and  $\text{Cu}^{2+}$  (Zhan et al., 2013), dithizone-immobilized zeolite for  $\text{Hg}^{2+}$  adsorption (Mudasir et al., 2016), poly(methacrylic acid)/iron-oxide-coated zeolite for removal of  $\text{Mn}^{2+}$ ,  $\text{Fe}^{2+}$  and  $\text{As}^{3+}$  (Pak et al., 2018), humic acid-immobilized surfactant-modified zeolite for the adsorption of  $\text{Cu}^{2+}$  (Lin et al., 2011), and polyacrylamide-zeolite composite modified by phytic acid for the adsorption of  $\text{UO}_2^{2+}$ ,  $\text{Tl}^+$ ,  $\text{Pb}^{2+}$ ,  $\text{Ra}^{2+}$ ,  $\text{Bi}^{3+}$  and  $\text{Ac}^{3+}$  (Şimşek and Ulusoy, 2004). The inorganic modifications comprehend mostly changes and procedures of coating, layering, or supporting metals on

zeolites, aiming to improve some specific adsorption sites towards contaminants removal (Alswat et al., 2016; Jiménez-Cedillo et al., 2011; Kazansky and Pidko, 2005; Luo et al., 2019; Medina-Ramirez et al., 2019; Onyango et al., 2003; Pahlavanzadeh and Motamedi, 2020; Qiu et al., 2018; Salam et al., 2020; Simsek et al., 2013; Venkata Ramana et al., 2013; Zhang et al., 2019f).

## 6.2. Organic molecules

### 6.2.1. Dyes

Yazdani et al. (2014) synthesized chitosan/feldspar bio-based beads and tested for the removal of Acid Black 1 dye from aquatic phases in 31 different batch experiments. The results showed uptake of  $19.85 \text{ mg g}^{-1}$  under the optimum conditions of pH 3, the temperature at  $15^\circ\text{C}$ , initial dye concentration of  $125 \text{ mg L}^{-1}$ , and an adsorbent dosage of  $2 \text{ g L}^{-1}$ , which was very close to the predicted maximum adsorption amount by the statistical model of  $21.63 \text{ mg g}^{-1}$ . The mathematical modeling on the dyes' adsorption behavior illustrated that the process followed Langmuir isotherm, as well as pseudo-second-order kinetics. Langmuir's maximum sorption capacity was found to be  $17.86 \text{ mg g}^{-1}$ . The thermodynamic parameters were evaluated and revealed that the adsorption process was exothermic and favorable (Yazdani et al., 2014).

Saiphaneendra et al. (2017) used hematite and magnetite nanoparticles to functionalize reduced graphene oxide sheets via a facile one-step co-precipitation technique. It has been postulated that both minerals' co-existence on the graphene sheet causes synergistic effects to enhance adsorption. For comparison purposes, the authors studied the adsorption behavior of pure graphene oxide, reduced graphene oxide, reduced graphene oxide/hematite composite (surface area =  $230 \text{ m}^2 \text{ g}^{-1}$ ), reduced graphene oxide/magnetite composite (surface area =  $231 \text{ m}^2 \text{ g}^{-1}$ ), and reduced graphene oxide/magnetite/hematite composite (surface area =  $216 \text{ m}^2 \text{ g}^{-1}$ ) against methylene blue adsorption. The adsorption kinetics was well described by the pseudo-second-order model, and Langmuir adsorption isotherm for the equilibrium adsorption behavior of rGO- $\text{Fe}_2\text{O}_3$ - $\text{Fe}_3\text{O}_4$ . The maximum adsorption capacity was determined to be  $72.8 \pm 2.7 \text{ mg g}^{-1}$  (Saiphaneendra et al., 2017). Kang et al. (2018) provided a novel approach to "waste eliminates waste" by employing ilmenite's acid leachate as the precursor in its structure tailoring. They reported that the final product's morphology and structure could be controlled, originating from nanoparticles, microcubes, rhombohedrons to the microsphere, by merely varying synthetic parameters. Due to its large surface area ( $152.3 \text{ m}^2 \text{ g}^{-1}$ ) and large functional groups,  $\text{Fe}_2\text{O}_3$  microspheres efficiently removed organic dyes from aqueous solutions. The maximum adsorption capacities obtained by Sips model ( $R^2 = 0.9820$ ) fit were 723.8, 150.7, and  $54.5 \text{ mg g}^{-1}$  for Congo red, Methyl orange, and Methylene blue, respectively, with  $0.3 \text{ g L}^{-1}$  of adsorbent, at pH of  $7.2 \pm 0.1$  and at  $25^\circ\text{C}$  (Kang et al., 2018).

Thermally treated, modified, and calcined dolomites have been used for dye adsorption. In 2010, Dolomite was thermally treated at  $1000^\circ\text{C}$ , going from a surface area of  $1.82$  to  $11.36 \text{ m}^2 \text{ g}^{-1}$ . The batch adsorption experiments were performed at  $25^\circ\text{C}$ ,  $0.06 \text{ g L}^{-1}$  of adsorbent, and aqueous orange I solution in a concentration range of  $20$ – $200 \text{ mg L}^{-1}$ . The synthesized material's final uptake was  $25 \text{ mg g}^{-1}$  (Boucif et al., 2010). In 2018, the adsorption of reactive black 5 and congo red by dolomite treated at  $900^\circ\text{C}$  was investigated in single and binary solutions. At equilibrium, congo red was more strongly coadsorbed than reactive black 5, with maximum adsorption capacities of 229.18 and  $72.37 \text{ mg g}^{-1}$  at  $40^\circ\text{C}$ , respectively (Ziane et al., 2018). At last, in 2019, low-cost porous calcined dolomite microspheres were prepared by simple spray drying and subsequent calcination. The material was tested against methylene blue removal in the conditions of  $20 \text{ g L}^{-1}$  of adsorbent and  $100 \text{ mg L}^{-1}$  of dye. The adsorption kinetics followed the pseudo-second-order, while the isotherm data fit the Langmuir model.

In this context, the microspheres' removal efficiency reached 95.6% (Yan et al., 2019).

Mahmoodi and Saffar-Dastgerdi (2019) prepared sodalite zeolite nanoparticles, subsequently modified by different amounts of 3-aminopropyl-triethoxy-silane, resulting in products with 2.3 wt.%, 4.4 wt.%, and 6.5 wt.% of the organic modifier, named 0.5, 1, and 1.5 zeolites, respectively. The surface-modified products were used for adsorption of Direct Red 23 and Direct Red 80 from simulated wastewater. After initial tests, 0.5 zeolite was elected as the best, and it was tested furtherly. Equilibrium data followed Langmuir isotherm and pseudo-second-order kinetic. The adsorption capacity of the 0.5 composite reached expressive 2415 and  $4842 \text{ mg g}^{-1}$  for Direct Red 80 and Direct Red 23, respectively. Thermodynamics was studied and measured Gibbs free energies at 298, 313, 323, and  $333 \text{ K}$  were  $-12.53$ ,  $-14.68$ ,  $-16.12$ , and  $-17.55 \text{ kJ mol}^{-1}$  for Direct Red 23, and  $-9.27$ ,  $-11.73$ ,  $-13.37$ , and  $-15.01 \text{ kJ mol}^{-1}$  for Direct Red 80, respectively. The adsorption process was physisorption, exothermic, and spontaneous (Mahmoodi and Saffar-Dastgerdi, 2019).

### 6.2.2. Pharmaceuticals

Martín et al. (2019) modified natural montmorillonite and a synthetic mica with cationic octadecyl-amine by a cation-exchange reaction and explored its potential use adsorbent for ibuprofen removal. The adsorption equilibrium isotherm was fitted with the Langmuir and Freundlich mathematical models ( $R^2 > 0.999$ ). The adsorption rate of  $\text{C}_{18}$ -montmorillonite (removal efficiency = 99.9%) was not dependent on ibuprofen concentration ( $0.1$ – $80 \text{ mg L}^{-1}$ ), but  $\text{C}_{18}$ -mica behaved contrarily (from 99.9% at  $0.1 \text{ mg L}^{-1}$  to 67% at  $80 \text{ mg L}^{-1}$ ). Variations in pH ranging from 4 to 9, did not affect the process. Pseudo-second-order kinetic model best described the adsorption of ibuprofen ( $R^2 > 0.993$ ), reaching equilibrium, with outstanding efficiency up to 100%, in less than 5 and 60 min for  $\text{C}_{18}$ -montmorillonite and  $\text{C}_{18}$ -mica, respectively (Martín et al., 2019).

Azizi (2020) studied a green preparation method of iron oxide/cellulose nanocomposite, used to remove metronidazole, an antibiotic, from aquatic solutions in the form of an aqueous extract of spent tea waste, regarding its effective and magnetic separation capability. The prepared nanocomposite had spherical particles with an average size of  $15.5 \text{ nm}$ . At pH 5, the metronidazole concentration of  $10 \text{ mg L}^{-1}$ , the adsorbent dosage of  $25 \text{ mg L}^{-1}$ , and contact time of 30 min, the pollutant's removal efficiency was 97.04%. Langmuir isotherm determined the maximum adsorption capacity of  $332 \text{ mg g}^{-1}$ , pseudo-first-order kinetic constant rate of  $1.15 \text{ L min}^{-1}$ , with the determination coefficient of 0.97 (Azizi, 2020).

The adsorption and advanced oxidation of diverse pharmaceuticals and personal care products from the water were studied by Masud et al. (2020). Interestingly, they started assuming that their model pollutants are found as a complex mixture in wastewater and the environment and used reduced graphene oxide as a support for nano-zerovalent iron against a complex mixture of 12 diverse personal care products (including antibiotic, anti-inflammatory, anti-seizure, and antidepressant pharmaceuticals). This approach is exciting for most studies on removing personal care products results with only one pollutant at a time, typically at high initial concentrations, not environmentally relevant. Considering starting concentration of  $200 \mu\text{g L}^{-1}$ , an adsorbent dosage of  $533 \text{ mg L}^{-1}$ , pH 3, and after a contact time of 30 min, final removal efficiencies for venlafaxine, citalopram, paroxetine, fluoxetine, diclofenac, ibuprofen, naproxen, acetaminophen, carbamazepine, lamotrigine, sulfamethoxazole, and caffeine reached 88.5%, 98.92%, 99.58%, 98.71%, 97.92%, 88.55%, 99.25%, 82.97%, 85.75%, 95.23%, 74.13% and 93.12%, respectively (Masud et al., 2020).

De Oliveira et al. (2020) investigated micro and mesoporous silica prepared with coal fly ash as starting material and used as an adsorbent to remove parabens. The composite prepared at pH 7 presented higher adsorption capacity, with a surface area of  $396 \text{ m}^2 \text{ g}^{-1}$ . Adsorption kinetics were carried out in a batch system with a 40 mL multicomponent

paraben solution at 5 mg L<sup>-1</sup>, 2 mg adsorbent mass, pH 3, agitation at 300 rpm for 30 min at room temperature (28 ± 2 °C). The same conditions were used in equilibrium tests, but the multicomponent parabens concentrations varied from 1 to 20 mg L<sup>-1</sup>. The adsorption capacity was determined as 0.01, 0.07, 0.54, and 1.31 mg g<sup>-1</sup> for methylparaben, ethylparaben, propylparaben, and butylparaben, respectively, by fitting the data obtained to Freundlich isotherm ( $R^2 > 0.999$ ) (De Oliveira et al., 2020).

Differently than the previous minerals, modified clays have been prepared by several different research groups and applied for the adsorption of many pharmaceuticals, such as amoxicillin (Pierucci et al., 2017), atenolol (Seema, 2013), carbamazepine (Dordio et al., 2009; Styszko et al., 2015), chloramphenicol (Lawal and Moodley, 2018), cinnamic acid (Calabrese et al., 2017), ciprofloxacin (Hamilton et al., 2014), diclofenac (Pierucci et al., 2017; Styszko et al., 2015), furosemide (Machado et al., 2017), gemfibrozil (Dordio et al., 2017), ibuprofen (Dordio et al., 2009; Styszko et al., 2015), ketoprofen (Styszko et al., 2015), mefenamic acid (Dordio et al., 2017), metronidazole (Calabrese et al., 2013; Kalhori et al., 2017), nalidixic acid (Lawal and Moodley, 2018), naproxen (Dordio et al., 2017), ofloxacin (Wang et al., 2014), oxytetracycline (Figueroa et al., 2004), promethazine (Gereli et al., 2006; Seki and Yurdakoç, 2009), propranolol (Pierucci et al., 2017), sulfamethoxazole (Lawal and Moodley, 2018), tetracycline (Figueroa et al., 2004; Lawal and Moodley, 2018; Liu et al., 2011), trimethoprim (Bekçi et al., 2006) and venlafaxine (Silva et al., 2018).

## 7. Conclusion

After exploring the literature, it was stated that unmodified and modified geological materials have yet been poorly explored as adsorbents. The studies performed report generally high adsorption ability against inorganic and organic pollutants, associated with low-cost in retrieving, recycling, and applying these materials for environmental remediation purposes. In general, all minerals presented interesting  $q_{max}$  values for metal ions adsorption, while clay composites presented themselves as a more suitable choice for removing dyes and other organic pollutants. Besides, geopolymers rise in the “from waste to waste” approach, re-destining environmental liabilities, such as fly ash, by using polymerization reactions between minerals containing aluminum and silicon and an activating agent. Overall, the geological originated materials represent promising candidates for removing metal ions, antibiotics, phenolic compounds, dyes, pesticides, and herbicides in their modified and unmodified versions, from various industrial wastewater types and aquatic environments.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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