Chapter 9
Remediation of Environmental Pollutants Using Nanoclays

Mohsen Soleimani and Nasibeh Amini

9.1 Introduction

Nanotechnology and nanomaterials have become more widespread for environmental pollution control over the last decades. Having complex properties, clay minerals are considered very useful in various environmental applications. The terms “nanoclays” and “clay minerals” are mostly used interchangeably because the clay minerals are particles with at least one dimension in the nanometer range (Zhu and Njuguna 2014). Nanoclays are layered silicates clay minerals which can be formed as a result of chemical weathering of other silicate minerals (Bignon 2013; Floody et al. 2009; Choy et al. 2007).Use of nanoclays is increasing in comparison to other nanomaterials because of their specific physicochemical properties (Zhu and Njuguna 2014). Nanoclays have widespread uses in various fields including food industries, agriculture, animal feed as well as environment remediation and medicine (Choy et al. 2007; Newman and Cragg 2007).

Since clay minerals have small particle size, high specific surface area (external and internal surface area up to 100 m^2/g), and a large porosity (Yuan and Wu 2007), they are suitable for pollution control and environmental protection among different nanomaterials. In addition, clay minerals are generally nontoxic and rather inexpensive sorbents (Churchman et al. 2006). Clays and clay minerals are interesting because of their common availability and extraordinary properties (Bergaya and Lagaly 2006). These valuable materials have been used in removal of pollutants from agricultural leachates, soil liners, and water and wastewaters because of their relevant properties mentioned in Table 9.1.

M. Soleimani (✉) • N. Amini
Department of Natural Resources, Isfahan University of Technology, Isfahan 84156-83111, Iran
e-mail: m.soleimani@cc.iut.ac.ir

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Table 9.1 Applications of clays for pollution control and environmental protection (Churchman et al. 2006)

Contaminants for control	Status (actual or potential use)	Relevant clay properties
Heavy metal cations and simple cations	Actual, mainly passive, use (e.g., in soils, liners)	Charge, surface area, reactive surface groups
Organic and biological cations	Potential for water and wastewater treatment, pesticide control	Charge, surface area, especially interlayer
Nonionic organic molecules	Actual, for water and wastewater treatment; potential, for pesticide control, waste liners	Charge, specific surface area, interlayer
Anions	Actual, for water and wastewater treatment; potential, for pesticide and nutrient leaching control	Charge, reactive surface groups
Turbidity and residual treatment chemicals	Actual, for treatment of potable water and some wastewaters and sewage	Colloidal, size and charge; surface area
Leachates	Actual, for waste liners and radioactive waste storage	Swelling, charge, surface area, reactive surface groups

Cation exchange capacity (CEC) of nanoclays is also a positive factor for sorption of various pollutants from contaminated environments (Churchman et al. 2006). This is particularly important for sorption and control of heavy metal cations which can occur at various sites on the surface of clay mineral particles (Inskeep and Baham 1983). Adsorption of heavy metal ions on surface of clay minerals is a complex process indicating formation of covalent bonds. The sorption of heavy metals does not solely depend on CEC of clay minerals. The reason is that heavy metal adsorption takes place due to different processes (Jackson 1998). Adsorption and desorption of heavy metals (e.g., Cu and Cd) were reported on the borders and interlayer sites of montmorillonite as a clay mineral (Undabeytia et al. 1998, 2002). For each type of metal ions, the selected site might depend on various factors such as pH, ionic strength, and the anions that are present in solution (Undabeytia et al. 2002). The heavy metals adsorption capacity of clay minerals is not the same. Furthermore, nanoclays can also react to different types of organic compounds. Since the majority of clay minerals consist of the negatively charged sorbents, they have a strong tendency to adsorb organic cations and they may be limited for adsorption of organic species which are bond to positive charges (Theng 1974).

Considering the importance of clay minerals/nanoclays in environmental issues, in this chapter we present their characteristics and their relevant application in various pollutants removal from contaminated air, water, and soil media.

9.2 Structure, Classification, and Characteristics of Clay Minerals

9.2.1 Structures of Clay Minerals

The unique structure of clay minerals is like the small plates which include several crystal sheets having a replicate atomic structure of alumina and silica sheets. The alumina sheets (i.e., octahedral) consist of a composition of six hydroxyls or oxygen enclosing a metal atom such as aluminum, iron, magnesium, or other atoms (Murray 2006). In the other hand, each silica sheet (i.e., tetrahedral) consists of four oxygen that is linked to adjacent tetrahedral by sharing three corners. The basic building framework of all clay minerals is the same and consists of the tetrahedral and octahedral sheets which linked together by specified ways and create a nano-structure. The fundamental unit of clay mineral particles consists of nanoparticles aluminosilicate with an outer diameter of 3.5–5.0 nm (Brigatti et al. 2006). The primary layers of smectite as a clay mineral have about 1 nm width and less than 100 nm length (Yuan 2004).

9.2.2 Clay Minerals Classification

The first classification of clay minerals at two-class (amorphous and crystalline) was proposed by Grim (1962). This classification becomes the basic of differences between various clay minerals (Murray 2000). Besides, the other classification of clay minerals is based on the layer type and charge per formula unit which has been shown in Table 9.2. In this classification, minerals which have sheets including 1 tetrahedral and 1 octahedral units are considered as 1:1 minerals and those have 2 tetrahedral and 1 octahedral units are 2:1 minerals. If the minerals of 2:1 type include a brucite layer ($Mg\ (OH)_2$), they may be categorized as 2:1:1 or 2:2 minerals. Furthermore, there is a simple classification of clay minerals in literatures which has classified them into four principal groups including illite, kaolinite, smectite, and vermiculite. The other classification is based on a scheme of hydrous phyllosilicate structures (Guggenheim et al. 2006).

9.2.3 Characteristics of Clay Minerals

The existence of negatively charged surfaces in the clay minerals is an important factor for CEC and swelling properties of the minerals. The charge in clay minerals is caused by their surface and structure. The surface charge usually depends on the value of environment pH while the structural charge originates from the interior of the layers and is permanent which is caused by ion exchange during the crystal

Table 9.2 Classification of clay minerals based on the layer type and charge per formula unit

Layer type	Group	Subgroup	Species
1:1	Kaolin-serpentine $x = 0$	Kaolin	Kaolin, dickite, nacrite, halloysite
		Serpentine	Chrysotile, lizardite, amesite
2:1	Pyrophyllite-talc $x = 0$	Pyrophyllite	Pyrophyllite
		Talc	Talc
	Smectite $x = 0.2–0.6$	Montmorillonite (tioctahedral smectite)	Montmorillonite, beidellite, nontronite
		Saponite (trioctahedral smectite)	Saponite, hectorite
	Vermiculite $x = 0.6–0.9$	dioctahedral vermiculite	dioctahedral vermiculite
		trioctahedral vermiculite	trioctahedral vermiculite
	Mica $x = 0.5–1.0$	dioctahedral mica	Muscovite, illite, glauconite, paragonite
		trioctahedral mica	Phlogopite, biotite, lepidolite
	Brittle mica $x = 2.0$	dioctahedral brittle mica	Margarite
		trioctahedral brittle mica	Clintonite, anandite
	Chlorite $x =$ variable	dioctahedral chlorite	Donbassite
		di- tri-octahedral chlorite	Cookeite, sudoite
		trioctahedral chlorite	Clinochlore, chamosite, nimite
	Palygorskite-sepiolite $x =$ variable	Sepiolite	Sepiolite
		Palygorskite	Palygorskite

Note x: charge per formula unit

formation. In the clay minerals of 2:1 layer type, the surface charge comes from fundamental surface of tetrahedral sheets, and in the clay minerals with 1:1 layer the surface charge exists from both of tetrahedral and octahedral sheets, but the surface charge in the two type clay (1:1 and 2:1) comes from the edges of the sheets (Eslinger and Pevear 1988). The most important characteristic of clay minerals is CEC which is influenced by total layer charge. Since the surface layer charge is affected by pH, usually CEC is measured at neutral pH (Eslinger and Pevear 1988). Ions and water molecules could be sorbed into the space between the sheets of 2:1 clay minerals, while that is impossible in 1:1 minerals in which the sheets are strongly bond together and there is no free space.

9.3 Removal of Pollutants Using Nanoclay Minerals

9.3.1 Gas Pollutants

Industrial activities are the main source of release of toxic gases into the atmosphere. The reduction of gaseous pollutants, particularly volatile organic compounds, using thermal oxidation is expensive due to the need of high amount of energy. In this regard, more efficient processes combined with absorption techniques can be developed to improve the efficiency of catalytic oxidation and finally gas removal (Pires and Pinto 2010). Many studies have shown the absorption of polar and non-polar gases and water vapor molecules by different types of nanoclay minerals, specially pillared nanoclays which have been widely developed for air pollutant abatement nowadays. Nanoclays can be interesting alternatives to reduce and finally remove gaseous pollutants due to having hydrophobic and hydrophilic properties. According to the wide use of nanoclays in control of gaseous pollutants, the researchers have improved sorption processes and control of gaseous pollutants using modified nanoclays. Molina-Sabio et al. (2004) investigated the adsorption of NH_3 and H_2S on activated carbon-sepiolite pellets. Nguyen-Thanh et al. (2005) have studied hydrogen sulfide adsorption using Na-bentonite modified by iron. The iron-doped samples revealed a significant increase in the clay absorption capacity and H_2S removal. Table 9.3 summarizes recent studies regarding the use of clay minerals as the sorbent of various gaseous pollutants. Furthermore, nanoclays have a high potential to remove air pollutants originated from various sources but their efficiency depends on the type of pollutants, the sorbent characteristics, and physicochemical environmental conditions. Besides, using the hybrid sorbents including nanoclays could be a good approach to enhance the pollutants removal efficiency.

9.3.2 Water Pollutants

Industrial, agricultural, and urban wastewaters contain high amounts of heavy metals and organic compounds. These pollutants have hazardous effects on human health and living organisms. Although there are many sorbents used in removal of water pollutants, nanoclays are evaluated as suitable sorbents because of their low cost and high removal efficiency. Therefore, clay minerals and nanoclays have widely used for water and wastewater treatment to remove heavy metals, organic pollutants, and nutrients from water resources (Abdelaal 2004). Clay minerals, such as bentonite and zeolite, having a large specific surface area and negative net charge can adsorb inorganic and organic cations from the environment (Konig et al. 2012; Babel and Kurniawan 2003; Murray 2000).Their capability in adsorption of Cr (VI) from a solution containing 5 ppm Cr and 0.4 g/L sorbent in batch experiments was more than montmorillonite and significantly enhanced when the sorbents were modified by TiO_2 (Fig. 9.1). It seems that we have to modify the

Table 9.3 Removal of various gaseous pollutants using different clays

Pollutant	Clay mineral	Removal (%)	Reference
p-xylene	Na-montmorillonite	–	Cabbar and Cakanyıldırım (2008)
n-Hexane	Montmorillonite		Morozov et al. (2014)
Benzene		–	
SO_2	Palygorskite	–	Zhang et al. (2009)
SO_2	Zeolite	>95 %	Vieira et al. (2011)
SO_2	Zeolite Clinoptilolite	>24 %	Allen et al. (2009)
SO_2	Zeolite Clinoptilolite	–	Ivanova and Koumanova (2009)
H_2S	Zeolite Clinoptilolite	–	Ozekmekci et al. (2015)
H_2S	Kaolinite	–	Batista et al. (2014)
H_2S	Bentonites	–	Stepova et al. (2009)
H_2S	Montmorillonite	–	Nguyen-Thanh et al. (2005)

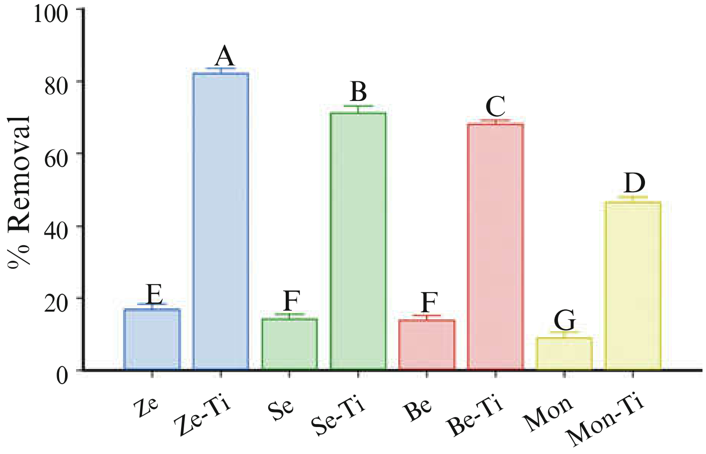

Fig. 9.1 Cr (VI) removal by four nanoclays (*Ze* zeolite, *Se* sepiolite, *Be* bentonite, *Mon* montmorillonite) and their modified types with titanium (Ti) in batch experiments (initial Cr concentration 5 ppm, sorbent dose: 4 g/L, pH: 2, titanium: 13 mmol Ti/g clay, Time: 90 min). The difference letters show the significant difference of means using Tukey test ($p < 0.05$)

clays in some cases in order to sorb a special pollutant and/or to increase their removal efficiency. Gu et al. (2010) compared adsorption of five different metal ions, Cd^{2+}, Cu^{2+}, Pb^{2+}, Ni^{2+}, and Zn^{2+}, by montmorillonite considering pH and ionic strength of the solution. Coppin et al. (2002) studied adsorption of lanthanide on kaolinite and Na-montmorillonite over a pH range and various ionic strengths. Adsorption of Pb onto sepiolite under different pH and temperatures was studied by Bektaş et al. (2004). Table 9.4 summarizes recent studies regarding the use of clay minerals as the sorbents for water pollutant removal.

9.3.3 Soil Pollutants

In recent years, soil pollution has significantly increased due to release of pollutants directly to the environment from various sources such as spills during

Table 9.4 Removal of water pollutants using various clays

Pollutant	Clay mineral	Percent removal	Reference
Cu (II)	Bentonite	48 %	Al-Qunaibit et al. (2005)
Cd (II)–CO(II)	Kaolinite	100 %	Angove et al. (1998)
Fe (II)	Bentonite	>98 %	Tahir and Rauf (2004)
Cu (II)–Zn (II)	Bentonite	99 % Zn (II) 90 % Cu (II)	Veli and Alyüz (2007)
Cd (II)	Montmorillonite	90.2 %	Undabeytia López et al. (1998)
Pb (II)	Zeolite clinoptilolite	–	Inglezakis et al. (2007)
Cu (II)- and Cd (II)	Na-montmorillonite	–	Inskeep and Baham (1983)
Cu (II)- and Cd (II)	Bentonite	84.5 % Cu (II) 87.2 % Cd (II)	Karapinar and Donat (2009)
NH_4	Zeolite clinoptilolite	>90 %	Rahmani et al. (2009)
Phenol	Bentonite	>45 %	Banat et al. (2000)

transportation, leakage from waste disposal or storage sites, and from industrial facilities (Riser-Roberts 1992). Therefore, soil pollution should be controlled via environmental friendly and low cost technologies (Riser-Roberts 1998). Clay is a significant vector transporting pollutants in the soil environment. It's a component of the soil with the ability to adsorb and transport compounds and nutrients, such as phosphorus (Wilkinson et al. 2000; Sharpley et al. 1984), potassium (Petrofanov 2012), metals (Quinton and Catt 2007), and organic pollutants (Wilkinson et al. 2000).

A major part of soil pollutants passes from the soil solution into the groundwater zone. Normally, different adsorbents exist in soil environment which among them clays with high CEC can bind to the positively charged organic materials and metals and make them chemically immobile and consequently reduce their imposed risk to the soil system. The value of soil CEC generally depends on organic substance and clay content of the soil (Pisani and Mirsal 2004). Sorption of ions and other compounds by clay minerals can be performed by two mechanisms: (1) adsorption on planar external surfaces and (2) exchange in the interlayer space. The presence of other minerals (e.g., Al and Fe hydroxides) may considerably increase CEC of clays such as montmorillonite (Terce and Calvet 1977).

Since, clay minerals consist of some silicates and organic components; they are very important sorbents in the soil environment. Due to the presence of clays in the soil, water molecules are adsorbed on their surfaces and this provides sorption sites for pollutants. Therefore, structural properties of individual clay minerals play a main role in determination of selectivity and adsorption mechanism of pollutants. As a consequence, the intensity of sorption in soils will mostly depend on the clay content and clay mineralogy in the soil texture (Pisani and Mirsal 2004).

Clays can be suspended in liquid phase of a porous media and also sorb the contaminants from the solution and therefore may be suitable for a safe infiltration process. Additionally, application of clay minerals in soil to keep the environment safe has been investigated in fundamental problem studies during last decades

(Kühnel 1990). Nowadays, nanoclays are used as a cover for landfills to prevent groundwater contamination. Application of clays in waste disposal as a buffer improved immobilization of hazardous compounds through isolation, stabilization, and fixing (Kühnel 1990). In other words, nanoclays and clay minerals in soil together with metal hydroxides and organic matter control the concentration of heavy metal ions in soil and groundwater. This depends on the type of heavy metal and solution pH and other environmental factors (Churchman et al. 2006). Pedogenic oxide is more effective than organic materials in removing Co^{2+} in the solution (McLaren et al. 1986). Furthermore, combinations of soil components and nanoclays organic matter complexes could enhance heavy metal adsorption. Many reports have mentioned the role of clays in soil stabilization in fields (Morgan 1995). Nanoclays have been used to remove polychlorinated biphenyl (PCB) from the soil (Mobasser and Taha 2013). The results clearly indicated that maximum percentage of PCB adsorption by nanoclays was 77 % (Mobasser and Taha 2013). The removal of diclofenac from soil using nanoclays revealed that the presence of clays in the soil could enhance the pollutant removal efficiency (Santin 2014). Adsorption of organic materials through weak Van der Waals forces on the surfaces of clay minerals can play a significant role in linking organic matter to the surfaces of clays (Stevenson 1994). Organic materials such as fulvic acid could be adsorbed into the space between the layers of montmorillonite (Schnitzer and Kodama 1977). Wei et al. (2015) investigated the removal of pyrene (i.e., one of the polycyclic aromatic hydrocarbons) by kaolin and montmorillonite (Wei et al. 2015). Their results showed that the removal of the pollutant by kaolin increased during the time while desorption from montmorillonite approximately remained stable during the time. The importance of clay minerals and organic matters in sorption of Triton X-100 confirmed the high potential of these compounds by montmorillonite (Zhu et al. 2003). Thus, the clays have a high potential to sorb, remove, and stabilize various pollutants in the soil media.

9.4 Conclusion

Clay minerals and nanoclays as the cheap and environmental friendly sorbents having unique characteristics (e.g., high surface area and porosity, mostly negative charged surface) can be used in various environmental issues. Considering the importance of control and remediation of environmental pollutants throughout the world, especially in developing countries, removal of air, water, and soil pollutants using nanoclays could be considered as a promising approach. Although clays have showed a high potential in removing or stabilizing various organic and inorganic pollutants in the environment, their efficiency depends on their type and structure as well as type of pollutants and the conditions of environment. The use of nanoclays with other sorbents or in combination with other remediation methods may be a promising approach to extend their capability in environmental management issues.

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