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Utilization and Environmental Concerns of $Al_2Si_2O_5(OH)_4$ and its Equilibrium Models in the Adsorption of Toxic Materials: A Review

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Abstract: Kaolin is an aluminum silicate material with the chemical formula $Al_2Si_2O_5(OH)_4$ with a 1:1 layered structure. It is a group of naturally occurring minerals, such as kaolinite and halloysite, formed from hydrothermal processes. Kaolin is classified into four different colors, each with its specific use. It contains silica and alumina compounds, making it a useful raw material for various applications. It is utilized as a raw material to manufacture paper, cosmetics, paints, etc. The expansion of the kaolin global market in big economic nations is due to the implementation of sustainable projects such as industrialization and urbanization. People in most parts of the world use kaolin as food and as a diet for some animals. In experimental laboratories, kaolin clay is used as an adsorbent to remove wastewater contaminants and protect municipal landfills from the effects of chemicals from solid wastes. The theories of various equilibrium models, such as Langmuir, Freundlich, etc., have been outlined in the removal or adsorption of hazardous materials using kaolinite clay as an adsorbent.

Keywords: *Kaolin, utilization, environment, silica, alumina, adsorption*

1. Introduction

The word kaolin originates from a hill in China called kao-ling, which simply means high ridge (Haq, *et al.*, 2008) The high ridge is referred to a hill in the South-Eastern part of China where the kaolin clay originated and was utilized to make porcelain centuries ago. The process and use of kaolin clay were duplicated after centuries later. Kaolinite is a dioctahedral phyllosilicate having a formula $Al_2O_3.2SiO_2.2H_2O$ or $Al_2Si_2O_5(OH)_4$ with a 1:1 layered structure of $[(Si_2O_5)]^{2-}$ and $[Al_2(OH)_4]^{2+}$ molecular sheets and composition of 39.5 wt % Al_2O_3, 46.5 wt % SiO_2 and 14 wt % H_2O (Alves et al., 2016; Sánchez-Soto, *et al.*, 2022).

Kaolins are rocks that comprised the kaolin group of minerals (kaolinite, halloysite, dickite, and nacrite), and they are formed both by weathering and hydrothermal alteration processes. It is a layered silicate mineral with a tetrahedral silica layer (SiO_4) bonded through oxygen atoms to an octahedral layer of alumina (AlO_6) (Kumari & Mohan, 2021). It is a white odorless naturally occurring aluminum silicate mineral (de Mesquita, Rodrigues, & Gomes, 1996; Zegeye et al., 2013). The white kaolin can also be formed by the removal of other colored compounds. In other parts of the world, kaolin is pink-orange-red and light concentrations produce white, yellow, or orange colors. Kaolinite is the principal component of the mineral and it is found alongside other minerals like quartz, feldspar, anatase, illite, mica, and muscovite (Parsons et al., 2003). Rocks rich in kaolinite are called kaolin (Nagasawa, 1978). It has a melting point of 740-1785°C and a density of 2.65 g/cm3. It is a major starting material that is used in manufacturing industries like paper, plastic, rubber, ceramic, drying agents, petroleum/petrochemical, construction material, chemical, pharmaceutical, and cosmetic products (Hosseini & Ahmadi, 2015; Patwardhan, 2013). Kaolin-coated paper is used in the manufacture of cigarettes.

The demand for polymer composites reinforced or filled with natural fibers and powders for various industrial applications (construction, automobiles, furniture, and sporting goods) is growing because they retain good environmental performance, low cost, and ease of production compared to traditional materials (Vedrtnam, 2018). Natural polymers as celluloses can also be used for removal heavy and pollutants from wastewaters (Errich, *et al.*, 2021; Akartasse, *et al.*, 2022; Manchala, *et al.*, 2022; El Hammri, *et al.*, 2022; Yang, *et al.*, 2024). They have low prices and low weight, and they minimize the impact on the environment; these are the key reasons for the rapid development of polymer composite materials. Recent research efforts are aimed at finding alternative fillers to replace inorganic fillers (Nishino, *et al.*, 2003).

Aluminum is a good engineering material with excellent properties and quality to form lightweight materials when combined with other elements to increase its strength and acquire other properties. The properties of aluminum justify its leading role for many engineering purposes and the potential increase of its global demand in the future (Beloglazov, *et al.*, 2012; Green, 2007). The scarcity of good-grade bauxite (Aluminum oxide) in Europe led to its price increase and was listed among Europe's critical raw materials for 2020 which is clear evidence of the need to use aluminum as an alternative material (Habert et al., 2020; Mathieux et al., 2017). Kaolinite is formed by the chemical erosion of rocks in a hot and humid climate such as rain forests (Bloodworth, *et al.*, 1993; Sánchez-Soto et al., 2022; Zegeye et al., 2013). The mineral clay formed from the hydrothermal weathering and volcanic eruption of rocks may be contaminated with harmful heavy metals and other impurities in different concentrations (Silva-Valenzuela et al., 2013). It is, therefore, necessary to purify the clay before application to ensure a desirable product (Silva-Valenzuela et al., 2013). The use of natural uncontrolled wastes in a variety of applications has gained worldwide attention due to industrialization and urbanization (Gündüzalp & Güven, 2016). This has led to the accumulation and utilization of waste (alumina and silica wastes) as raw materials to manufacture new products for safe human health and environmental pollution and contributes to economic development (Yagci, *et al.*, 2006). Considerable human exposure is encountered during the mining and refining of kaolin and in its applications. Staying close to kaolin for a long period causes radiological complications and leads to reduced respiratory function (Parsons et al., 2003; Pope III, 2000). Studies have shown that exposure to the quartz contained in kaolin will lead to lung cancer and silicosis, and continuous exposure causes chronic bronchitis and pulmonary emphysema. Kaolin is released into the environment during its use in domestic, paper, petroleum, construction materials, etc. It is also directly released into the environment during the formulation of insecticides. This work reviews kaolin utilization and its associated environmental health concerns. Mine workers are exposed to kaolin dust and it has been reported from other studies that people are exposed to kaolin through consumer products containing kaolin.

1.2. Classification of kaolin natural colors and uses

Figure 1 shows the different natural colors of kaolin clay. White kaolin is the mildest clay among other types of clays and comprises higher quantities of silicon and aluminum silicate materials. It is sensitive to the skin and is often used for soaps and skin. Yellow kaolin consists of titanium, potassium, and silicon with improved soft and mild efficacy. It is safe for dry, sensitive, and normal skin. Red kaolin comprises higher components of iron oxide and copper but has low aluminum content. Traditionally, red kaolin was utilized to treat joint pains and as a washing agent for infection and disease protection. Pink kaolin is a combination of red and white clay and has a high content of aluminum oxide. Pink clay is good for normal skin. It has a combined property since it is a blend of both red and white clay. It fights skin dryness and irritation. It absorbs toxins and clogged oil from the skin.

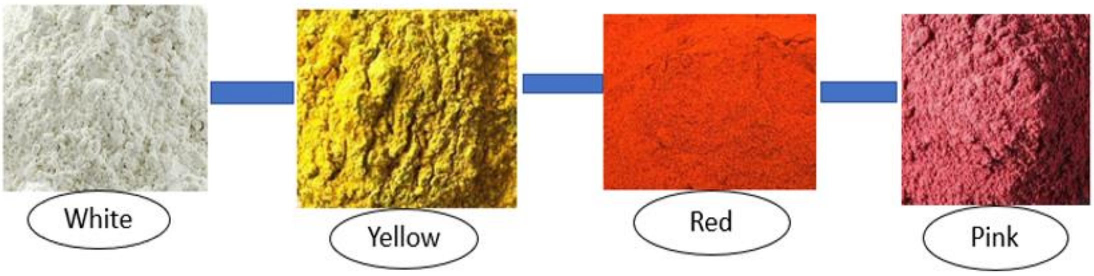

Figure 1. Classification of kaolin based on natural color

1.2. Kaolin market

The high demand for kaolin by the end user (paper, ceramics, sanitaryware, etc.) is significantly moving forward in the global kaolin market. The demand for ceramic tiles from highly emerging and populated nations might be a leading factor in the growth of the kaolin market. A report from Fortune Business Insights indicates that the kaolin market size is projected to rise to USD 5.7 billion in 2029. Kaolin is an important raw ingredient in the production of ceramic, tableware, tiles, big surfaces, electrical porcelain, etc (Štyriaková & Štyriak, 2000). The increase in the use of paper also influences the growth of the kaolin market. The pure and extremely powerful color pigment kaolin is used as coatings on the surface of papers to give a bright, silky, and glossy appearance, easier printing, ink adhesion, etc., of the paper. The use of kaolin in industries is increasing and is expected to contribute to the kaolin growth market over the forecast period. The demand for kaolinite clay in the Asia Pacific has grown as a result of economic expansion, urbanization, and industrialization. Japan, China, and India are the highest paper manufacturers with China having an annual production of 99.3 million tons. However, the identification of other materials to replace kaolin as raw material in the paper industry is a challenge for the growth of the kaolin market. The use of computer technology to replace conventional notebooks or paper, and regulations to control the cutting down of trees for kaolin mining are all factors that hinder the expansion of the kaolin ash market.

2. Utilization of kaolin

Kaolin exploitation has acquired great economic importance nowadays because of its applications in manufacturing products like paper, paints, plastics, etc (Ekosse, 2010). Kaolin has been utilized for several decades in various applications such as ceramics, cosmetics, agriculture, etc. Extensive research on the usage and toxicity of kaolin shows that kaolin is safe. However, non-pesticidal use of kaolin and exposure to and inhalation of dust particles by occupationally exposed workers in kaolin mining areas may lead to some complications.

2.1. Ceramics

Kaolin is utilized to manufacture ceramics in ceramic industries (Dewi, Agusnar, & Alfian, 2018; Ombaka, 2016; Sánchez-Soto et al., 2022). Ceramic and whiteware production is one of the most common and important applications of kaolin due to its ideal properties. Kaolin, silica, feldspar, and ball clay are mixed by ceramic producers to achieve plasticity, shrinkage, and vitrification to achieve proper shape. Kaolin is an important raw material employed in the manufacture of ceramics due to its high-temperature stability, high refractoriness, high resistance to corrosive agents, etc (Alves et al., 2016; Chakraborty, 2014; Chakraborty & Chakraborty, 2014). The mineralogical compositions of clays are important aspects to evaluate since they directly influence the physical and mechanical qualities of fabricated ceramic products (Azevedo et al., 2018; Gonçalves et al., 2017; Schroeder & Erickson, 2014).

2.2. Concrete and cement

The cement industry is one of the highest global energy users, and it is the second-largest carbon dioxide (CO_2) emitter and takes an 8–10% share of global emissions (da Silva, Malacarne, Longhi, & Kirchheim, 2021; ECRA & CSI, 2017; John, Quattrone, Abrao, & Cardoso, 2019; Miller, 2018; UNEnvironment, John, & Gartner, 2018). Kaolin is a popular additive product to use in concrete mixtures because it is a silicate mineral, which is necessary for giving concrete its required properties. Its demand in construction is rising because of its ability to provide bright white coloring to cement mixtures. As a concrete mix additive, kaolin is also ideal because it is chemically inert and will have minimal effect on the formula's chemical or physical properties. Kaolin is also required in cement because of its high activity which is its ability to react with Portland cement to accelerate the hydration of the Portland cement (Skibsted & Snellings, 2019).

2.3. Paper

Paper surfaces are coated to provide a sharp and bright printed photographic color. The most important properties of kaolin to the paper industry are whiteness, low viscosity, non-abrasiveness, etc. The white clay mineral augments the brilliant, glossy, opaque, etc appearance of the paper. It increases other paper qualities, such as tincture absorption, pigment holdouts, and low inclination to lint and roughness, thereby improving paper printability. The whiteness of a paper is one factor that mainly determines its market price.

2.4. Polymeric

Kaolin is utilized as a filler in plastic industries. It was at first used as a cheap raw material and processing aid in the production of polyvinyl chloride and polyesters.

Kaolin is used in polymers (plastics and rubbers) to improve smoothness, strength, stability, and resistance to chemical attack, resistance to tear, cracking, and abrasion, and reduce shrinkage during polymerization. It also improves these products' tensile strength and prevents warping. Kaolin also helps extend pigment and produce sharper colors in polymers, while helping to enhance the final products' smoothness and brightness. It acts as a reinforcer to improve the durability and rigidity of plastics and rubber.

2.5. Fiberglass

Kaolin is used to manufacture fiberglass which acts as a strength in many applications. Kaolin also promotes the integration of fibers to strengthen the plastics and rubbers used in cars, boats, marine products, aviation, and aerospace products, circuit board manufacturing, etc. It provides alumina, which is necessary for improving the durability of the product surface. Kaolin is utilized by manufacturers to improve quality, and modification, promote cost-effectiveness and increase the production of their products.

2.6. Agriculture

Kaolin is used in agriculture to protect crops and seed coatings from damage by insects and pest diseases. A solution of kaolin sprinkled on the surfaces of crops acts as a shield for leaves and fruits (Dinis et al., 2016; Gebremariam, Welde, & Kahsay, 2018). This means it can be used as a solar protectant to act as a shield or barrier to reduce exposure to heat and light thereby reducing sunburn of crops and enhancing photosynthesis. Seeds coated with kaolin are proven to have better germination and unfavorable climatic conditions. This technique ameliorates the loss of excess water in plants (Dinis et al., 2016). There are no associated health issues with eating food protected with a solution of kaolin. Kaolin application on apple trees maintained the structure of photosystem II, increased the net photosynthesis rate, reduced water consumption, reduced insect damage, controlled diseases, reduced frost damage, and increased the anthocyanin content of apple fruits (Glenn, Erez, Puterka, & Gundrum, 2003; Wand, Theron, Ackerman, & Marais, 2006). The use of kaolinite adequately solves the negative impacts of environmental conditions on crops and that subsequently leads to a high yield of agricultural products (Brito et al., 2018). Kaolin is utilized as a beneficial antitranspirant (Cantore, Pace, & Albrizio, 2009). It enhances the adequate use of water in several farm plants (Abdallah, El-Bassiouny, & AbouSeeda, 2019; Cantore et al., 2009; Ćosić et al., 2015; Ibrahim & Selim, 2010; Javan, Tajbakhsh, & Mandoulakani, 2013; Rosati, Metcalf, Buchner, Fulton, & Lampinen, 2006). In summary, farmers use kaolin to protect fruits and vegetables from the destruction of insects, fungi, bacteria, mites, heat, and sunburn.

2.7. Adhesives

Kaolin is used in adhesives to modify rheology enhance wet strength, whiteness, film strength, and durability, and reduce viscosity and shrinkage to enhance cost-effectiveness. The natural particle size and shape of kaolin make it a useful unique material in adhesives. Kaolin and white clay have the potential to be utilized as a geopolymer coating for application in pipeline systems. Studies have been done on the application of coatings to improve the technical performance in materials such as aircraft, marine, concrete, pipeline system, etc (Balaguru, Kurtz, & Rudolph, 1997; Bierwagen & Tallman, 2001; Zhang, Yao, & Wang, 2012; Zhang, Yao, & Zhu, 2010). The use of organic polymers as protective coatings is not user-friendly and hence detrimental to our health and environment. The adhesive action of pressure-sensitive styrene-diene and the viscoelastic properties of pressure-sensitive natural rubber using acrylic resin as a tackifier has been studied (Gerard Kraus & Hashimoto, 1982; G Kraus & Rollmann, 1977; Leong, Lee, & Gan, 2003). Coumarone-diene resin has also been used as a tackifier resin to study the adhesive properties of pressure-sensitive natural rubber (Poh & Chang, 2006; Poh & Chee, 2007; Poh & Kwo, 2007). Fillers are additives used to reduce cost and add to the body of adhesive or reduce the over-penetration into the wood, increase the rigidity of cured adhesives, and to also modify the thermal expansion of a film to approximately that of the adjacent adherence (Cao, Zhou, & Du, 2020; Dunky, 2003). The thermal conductivity of adhesives can be improved using copper powder, aluminum powder, etc, and they can be widely used in microelectronic assembly and bonding electronic products instead of spot welding(Cao et al., 2020).

2.8 Paints and coatings

Various types of mineral fillers such as kaolin, talc, etc derived from diverse geological formations are globally utilized (da Conceição, Petter, & Sampaio, 2018). Kaolin is used to improve whiteness, brightness, opacity, scrub resistance, and corrosion. The final products of kaolin paints offer better performance when painted on surfaces and hence increase its demand in the pain industry. Kaolin is used to replace the use of TiO_2 to manufacture paint with less environmental impact and improve the inconsistency in paint finish (Ninness, Bousfield, & Tripp, 2003; Vickers, 2017). It is a well-known and valuable mineral filler with fine-grained size and plate-like aluminosilicate particles making it very easy to substitute TiO_2 in pain formulations (Ahmed, 2013). Fillers or binders are finely-grained solids used as mineral pigments added to polymers for quality enhancement (Gaikwad & Ko, 2015). They are finely ground powders, do not dissolve in the application medium, and are purposely utilized to beautify and increase the quality of the product (Svyatchenko, Kiryushina, & Sharapov, 2020; Vickers, 2017).

2.9 Wastewater treatment

Kaolin is utilized as an adsorbent in water and wastewater treatment. Kaolin products are affordable and can be used as an alternative for the treatment of wastewater minerals. It is an effective material for removing wastewater toxins and contaminants such as chloride and iron. Wastewater treatment can be done using various methods such as filtration, oxidation and evaporation, centrifugation, flotation, distillation, ion exchange, precipitation, adsorption electrolysis, electrodialysis, sedimentation, gravity separation, reverse osmosis, coagulation, etc (Cardenas et al., 2016; da Conceição et al., 2018; Huang et al., 2016; Li et al., 2016; Mousa & Hadi, 2016; Peeters, 2015; Sun et al., 2017; Tan, Huang, Wu, & Chen, 2017)

2.10. Pharmaceuticals

Kaolin is sometimes used to make medicine (Dewi et al., 2018). It can be used to stop bleeding, swelling, and mouth sores (mucositis) and decrease the pain in the mouth. Kaolin is also used to treat diarrhea and other conditions. Kaolin clay also has medicinal and therapeutic values that are useful for the body. It is the oldest ingredient used in various treatments depending on the mineral content of the clay. These various types of kaolin clay have been used as an ingredient for all-natural skincare items (Olaremu, 2015). Natural pure kaolin is used in the manufacture of drugs and cosmetics mainly as inert charges and also as an active constituent for cosmetic muds and stomach disorder medications (López-Galindo, Viseras, & Cerezo, 2007; Silva-Valenzuela et al., 2013). Finely divided particle-size kaolin with a large surface area that absorbs various compounds is used for pharmaceutical purposes. Kaolin agglutination is used in the serodiagnosis of tuberculosis (Sarnaik, Sharma, Kate, & Jindal, 1993). In animal husbandry, pectin is mainly used as an oral anti-diarrheal veterinary medicine. It is also employed as an adsorbent to cure people who mistakenly consume toxic materials (Gushit, Olotu, Maikudi, & Gyang, 2010). Despite the benefits we derive from the use of kaolin, we still need great care when using it for skin treatment purposes. Some grades are used for skin care treatment and others are used for industrial purposes. Industrial-grade kaolin is often mixed with other ingredients and it should therefore not be used for skin care.

2.11 Catalysts

Kaolin is the most important mineral for the manufacture of carriers for catalysts. Kaolin is largely utilized as a catalyst substrate in the catalytic cracking of petroleum. The refractory character of kaolin is suitable for this application at high temperatures and pressure. Purified kaolin is used in the catalytic cracking of petroleum. Kaolin is converted to zeolite to increase the surface area of the catalyst exposed in the reaction. It is a preferred catalyst carrier due to its cheap price, zeolite formation, high purity, etc (Hettinger Jr, 1991). It is estimated that over 200,000 tons of kaolin are used annually to produce petroleum-cracking catalysts (Hettinger Jr, 1991).

3. Relating kaolin to the environment and human health

3.1. Environment

Raw mineral materials like clays, gravel, etc., are often utilized for building, agriculture, and mitigation of environmental problems. This has prompted regulatory agencies for land management to have better geologic and mineral data on industrial minerals. Emphasis is laid on the characteristics of the life cycle of the clay deposit to help develop the geologic and geochemical information and also to determine the environmental problems that arise from the use of clays with the potential to affect its natural and industrial applications. Environmental characteristics include the nature and distribution of inorganic contaminants, such as metals and metalloids like arsenic, iron, and lead, in clay-bearing rocks. These environmental factors have the potential to affect the use of clays in natural and industrial applications. There is an increasing global concern about toxic environmental pollutants due to their severe health and environmental effects such as water pollution. Polluted water is a severe health problem for people due to contaminants from natural and anthropogenic sources. In most of the globe, toxic materials from several industrial practices are deposited into nearby water sources (Awaleh & Soubaneh, 2014). For instance, the wastewater from ternary industries is polluted with organic and inorganic compounds (Saxena, Chandra, & Bharagava, 2017). The wastewater is purified using some techniques like flotation, oxidation and evaporation, distillation, precipitation, electrolysis, adsorption, crystallization, micro and ultra-filtration, coagulation, etc (Mustapha et al., 2019). Kaolinite is a cheap adsorbent used to remedy disposal and wastewater purification problems and does not need to be regenerated (Uddin, 2017). It possesses high chemical stability, low expansion, and cation exchange capacity.

3.2. Protect landfills from solid waste chemicals

Toxic organic and inorganic municipal solid wastes are mostly generated from industries and contain cations due to the presence of salt (Boopathy, Karthikeyan, Mandal, & Sekaran, 2013; Colman, Bulteel, Rémond, Zhao, & Courard, 2020; Ozgunay, Colak, Mutlu, & Akyuz, 2007; Turkyilmaz et al., 2019; B. Wang, Xu, Chen, Dong, & Dou, 2019; Weber, Jang, Townsend, & Laux, 2002; Yeheyis, Hewage, Alam, Eskicioglu, & Sadiq, 2013). A decrease in osmotic potential will augment the hydraulic action of various inorganic salts (Lee & Shackelford, 2005; Setz, Tian, Benson, & Bradshaw, 2017; Tian, Likos, & Benson, 2019; Wireko & Abichou, 2021). The deposition of the inorganic material is inhibited using liners made of kaolin. The ionic salts provide smooth mobilization of microorganisms into the environment. Toxic organic and inorganic municipal solid wastes are mostly generated from industries and contain cations due to the presence of salt (Boopathy et al., 2013; Colman et al., 2020; Ozgunay et al., 2007; Turkyilmaz et al., 2019; B. Wang et al., 2019; Weber et al., 2002; Yeheyis et al., 2013). A decrease in osmotic potential will augment the hydraulic action of various inorganic salts (Lee & Shackelford, 2005; Setz et al., 2017; Tian et al., 2019; Wireko & Abichou, 2021)· The deposition of the inorganic material is inhibited using liners made of kaolin. The ionic salts provide smooth mobilization of microorganisms into the environment. The ionic salts provide smooth mobilization of microorganisms into the environment as demonstrated in **figure 2** below.

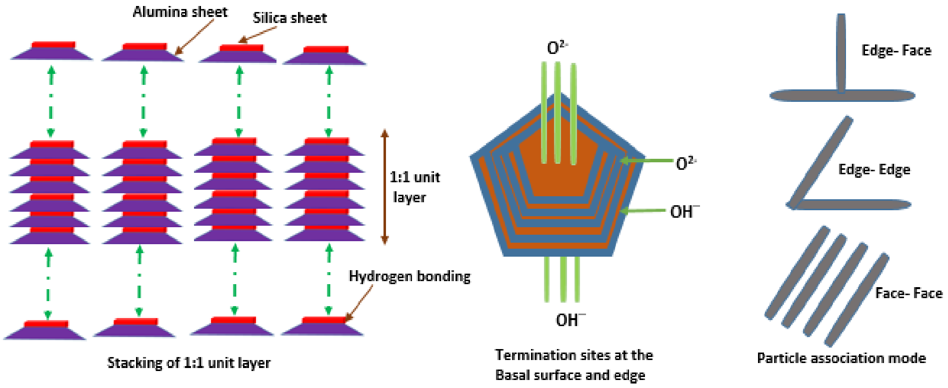

Figure 2. Occupational exposures to kaolin minerals

It has been speculated that cardiopulmonary illness in the elderly and children are the result of short-term exposure to exposure to PM10 (Pope III, 2000). Some researchers have investigated the toxicity of geologic materials such as those from the extraction of Chinese clay (Richards & Hunt, 1983). The doses are more applicable to occupational exposures and not exposures to environmental dust. Occupationally-related pneumoconiosis was first described in 1956 in china clay workers, and since then there have been several epidemiological studies on respiratory health and the china clay industry (Hale, Gough, King, & Nagelschmidt, 1956; Parsons et al., 2003). Studies have also considered occupational exposures to dust particles, radiological and pathological data, and internal dust accumulations(Lapenas, Gale, Kennedy, Rawlings Jr, & Dietrich, 1984; Oldham, 1983; Rundle, Sugar, & Ogle, 1993; Sheers, 1989). The inhalation of kaolin has been associated with many complications such as pneumoconiosis, kaolinite pneumoconiosis, or kaolinizes. Investigations have also been done on the epidemiology of china clay workers in Georgia (USA) (Lapenas et al., 1984). The toxicity of quartz in the lung is well established and commonly used as the ‘positive control’ in toxicity assays (Reynolds, Jones, BéruBé, Wise, & Richards, 2003; Richardson-Boedler, 2007).

3.4. The use of kaolin as a diet feed

3.4.1. Decontamination

Kaolin is adsorbent and primarily not toxic; it can therefore adequately be utilized to inhibit the severity of toxic substances both in living things and the environment. A combination of kaolin and pectin can be taken to mitigate diarrhea and digestive problems(Gebesh, Ianchenko, & IuA, 1999; Heimann, 1984). It has been validated by various studies that kaolin is capable of decontaminating aflatoxins, diarrhea-causing enterotoxins, pathogenic microorganisms, heavy metals, and poisons (Abdel-Wahhab, *et al.*, 1999; Berraouan *et al.*, 2020). According to Hashsham et al (Hashsham & Freedman, 2003), the adsorption of vitamin B12 is very slow in kaolin clay materials. In conclusion, when kaolin is ingested through diet, the harmful dietary compounds stick to the intestinal walls of the animals and retards their absorption through the intestinal mucus to stop their harmful acts on the animals.

3.4.2. Feeding

Research on the use of kaolin as nutrition for agricultural animals is limited. Savory (Savory, 1984) researched to investigate kaolin as a nutrition to feed adult roosters. He did not discover a change in body weight when he applied 100 and 200g kaolin/kg of the diet. The animals at first ingested 300 g kaolin/kg but there was no enlargement in their body weight. The weight depreciated when they consumed 400 g of the clay. This was noted in animals fed with 100 g kaolinite in a few days and when more than 200 and 300 g of kaolin per week and 400 g per three weeks, were consumed, respectively. The stimulation of the live weight gain in rats given kaolin combined with the diet (100 mg/g) with a concurrent proportional increase in the weight of the tissues of some digestive organs was reported by Sakata (Sakata, 1986).

3.4.3. Geophagy

Geophagy is observed in animals and the human population and it is sometimes deemed an adaptable way of living by other primates (Geissler, Mwaniki, Thiong'o, Michaelsen, & Friis, 1998; Johns & Duquette, 1991; Knezevich, 1998; Mahaney, Bezada, Hancock, Aufreiter, & Pérez, 1996). It is the habit of eating kaolin either for pleasure or to suppress hunger. Kaolin consumption is more prominent among women, particularly pregnant women (Geissler, Shulman, et al., 1998; Prince, Luoba, Adhiambo, Ng'uono, & Geissler, 1999; Sapunar & Fardella B, 1999). The use of kaolin by women in some African nations is sometimes equated to cigarette smoking habits by men. Animals and some human beings are generally adapted to the practice of eating kaolin (Dominy, Davoust, & Minekus, 2004). The absorption of harmful materials from plant products is decreased in birds practicing geophagy behavior (Diamond, Bishop, & Gilardi, 1999). Animals like rats that suffer from gastrointestinal disorders, arthritis, and stress are also involved in this practice (Morita, Takeda, Kubo, & Matsunaga, 1988). Vomiting appears to be a functionally similar defense behavior in humans and other animals (Takeda, Hasegawa, Morita, & Matsunaga, 1993).

3.4.4. Toxicity in animal

The intake of kaolin has not shown any toxic effects on organisms but it however becomes harmful when contaminated with dioxins. Contaminated poultry food samples with polychlorinated dibenzodioxins and polychlorinated dibenzofurans were detected in USA and Europe (Eljarrat, Caixach, & Rivera, 2002; Takeda et al., 1993) in which kaolin mineral was the source of contamination. It is important to take note of the factors that lead to the contamination of kaolinite clay (Matlova et al., 2004). Particulate matter and the soil above the kaolin layer can also pollute the kaolin. Animals like pigs are susceptible to mycobacterial infections like tuberculosis from *M. a. hominissuis*, *M. fortuitum, etc* (Matlova et al., 2012). Members of the *M. avium* complex (*M. a. avium*, *M. a. hominissuis*, and *M. intracellulare*) cause serious infections in animal and human populations (Bartos, Falkinham, & Pavlik, 2004; Dvorska et al., 2004; Lescenko et al., 2003; Machackova et al., 2003; Pavlik et al., 2004).

3.5. Application of mathematical models in the adsorption of toxic materials using kaolin.

3.5.1. Langmuir isotherm

Adsorption equilibrium provides a piece of comprehensive knowledge about the process of adsorption. It concerns the amount of solute on the adsorbed surface to the concentration of the liquid phase solute at equilibrium. The adsorption equilibrium capacity of the kaolin material in this work can be explained by isotherms. Initially, most isotherms were used for gaseous phase adsorption but they are nowadays utilized to reciprocate the adsorption equilibria of multiple substances including kaolin clay minerals. The equilibrium assimilation principles of toxic or hazardous materials in kaolin clay minerals can be complemented by applying the Langmuir and Freundlich equations (Ong et al., 2014; Yesi, Sisnandy, Ju, Soetaredjo, & Ismadji, 2010; Zhou et al., 2012). In 1918, Langmuir projected the theory for the assimilation of materials on surfaces assuming that the surface of the adsorbent is uniform, adsorption is confined on the surface and only one molecule is adapted to each active site on the surface of the adsorbent (kaolin clay). It has been proven that the rate of adsorption onto a flat surface equals the rate of desorption from the surface to maintain the rate of accumulation at zero at the surface at equilibrium conditions (Do, 1998). The expression below is the Langmuir equation for the adsorption of a single molecule of toxic substances (one molecule) on the active surface of the adsorbent:

$$q_e = q_{max} \frac{K_L C_e}{1+K_L C_e} \qquad (1)$$

Where:

q_e is the amount of the solute adsorbed onto the adsorbent surface at equilibrium condition,

q_{max} is the adsorption capacity of an adsorbent, while

K_L and C_e are affinity constant and the equilibrium concentration of solute in the bulk liquid, respectively.

The parameter K_L measures how strongly an adsorbate molecule is attracted to a surface(Do, 1998). Theoretically, when the value of the parameter K_L is larger, the interaction between the surface of the adsorbent and the solute molecules becomes stronger, and the surface of the adsorbent will be covered more with the solute molecules. One of the most popular models to determine liquid adsorption equilibria data is the Langmuir equation. The nonlinear regression rule is used to determine the parameters (q_{max} and K_L) of the Langmuir equation. The linear forms of the Langmuir equation can be written as follows:

$$\frac{C_e}{q_e} = \frac{1}{q_{max}} C_e + \frac{1}{K_L q_{max}} \qquad (2)$$

$$\frac{1}{q_e} = \left(\frac{1}{K_L q_{max}}\right) C_e + \frac{1}{q_{max}} \qquad (3)$$

$$q_e = q_{max} - \left(\frac{1}{K_L}\right) \frac{q_e}{C_e} \qquad (4)$$

$$\frac{q_e}{C_e} = K_L q_{max} - K_L q_e \qquad (5)$$

The dimensionless constant called the equilibrium parameter, R_L accounts for the existing characteristics of the Langmuir model (Kurniawan et al., 2011) written below

$$R_L = \frac{1}{1+K_L C_o} \qquad (6)$$

Where C_o is the initial solute concentration in the liquid phase. The classification of parameter R_L as the nature of adsorption is as follows: favorable convex isotherms ($0 < R_L < 1$), unfavorable convex isotherms ($R_L > 1$), linear (R=1), or highly favorable/non-reversible ($R_L = 0$).

3.5.2 Freundlich isotherm

This is a well-known adsorption model that accounts for equilibrium in the liquid phase. It is among the oldest hypothetical equations used to depict adsorption data in equilibria (Do, 1998). The adsorption of toxic liquid material in kaolin clay below:

$$q_e = K_F C_e^{1/n} \quad (7)$$

Where K_F and n are Freundlich parameters. The parameter K_F denotes the adsorption efficiency of the adsorbent, while parameter n shows that the system is heterogeneous.
The mathematical linear logarithmic model of Freundlich is expressed by the function below:

$$\text{In}q_e = \text{In}K_F + \frac{1}{n}\text{In}C_e \quad (8)$$

This model is suitable in most cases for both data from adsorption equilibria and heterogeneous systems (Chen & Liu, 2014; W. Wang et al., 2006). It lacks the adequate behavior of Henry's law when the concentration of a system is low, it is therefore only applied in solutes having a narrow range of concentrations and it does not have a finite limit when there is enough or high solute concentration (Do, 1998).

3.5.3 Hazardous chemical models

Gaseous phase assimilation models were employed to reciprocate the equilibria data for diverse hazardous substances on the surface of clay and kaolinite. The two fitting parameter models or equations are among these hazardous isotherms and they include Temkin isotherm, Flory– Huggins, BET equation, and Dubinin–Raduskevich equations. The sections below dilate the application of these isotherm equations to correlate the adsorption of experimental data of hazardous materials in kaolinite clay mineral systems.

3.5.4. Dubinin Raduskevich equation

Dubinin and his colleagues presented this semi-empirical model for microporous sub-critical solids vapors using a pore-filling mechanism for the adsorption process, where a pore-filling mechanism is used by the absorption process (Do, 1998). The Dubinin-Radushkevich equation for the assimilation of kaolinite clay is written as:

$$q_e = q_{max}\exp\left(-\left(\frac{RT\text{In}(C_e/C_S)}{\beta E_o}\right)^2\right) \quad (9)$$

Where:
β is the affinity coefficient for the volume of the liquid.
E_o represents the characteristic energy, while
C_s is the solubility of the solute at a given temperature.

The Dubunin-Radushkevich equation is not commonly used for liquid phase assimilation of harmful substances due to complicated factors such as pH and ionic equilibria that are present in such systems (Ismadji & Bhatia, 2001). This model has been used in many research to correlate the assimilation of equilibria data of kaolinite clay adsorbent and harmful materials (Hajjaji & El Arfaoui, 2009; Kadu & Chikate, 2013; Randelovic, Zhang, Jacimovic, McCarthy, & Deletic, 2016)

3.5.4.1. Temkin isotherm

The correlation of liquid phase assimilation of experimental data of dangerous substances and kaolinite is always reciprocated by using Temkin isotherm (Ahmedzeki, Rashid, Alnaama, Alhasani, & Abdulhussain, 2013; Gao et al., 2013). The Temkin model was developed assuming that the decrease in heat of sorption is more linear than logarithmic. The expression for the Temkin isotherm model is written as:

$$q_e = \frac{RT}{b_T}\text{In}(A_T C_e) \quad (10)$$

where A_T and b_T are equilibrium binding constant and parameter constant, respectively. The Temkin equation does not have the correct Henry law limit and finite saturation limit, it is therefore similar to the Freundlich model. The Temkin isotherm is not adequate to represent the adsorption equilibria data of hazardous substances and kaolinite clay minerals. It also fails to represent the adsorption equilibria data for complex liquid phase systems.

3.5.4.2. Flory Huggins isotherms

Unlike other models, this isotherm model is not always used to reciprocate the equilibria data assimilation of dangerous substances in the kaolinite system. It is employed to determine the area on the surface of the adsorbent covered by the solute. It also shows the feasibility and spontaneous disposition of the adsorption process. The Flory–Huggins isotherm equation is written as:

$$\frac{\theta}{C_O} K_{FH}(1-\theta)^{nFH} \qquad (11)$$

where θ is the degree of surface coverage and is defined as:

$$\theta = 1 - {C_e}/{C_O} \qquad (12)$$

Where: K_{FH} *and* nFH are Flory–Huggins isotherm model exponent (also correlates to the numberof solute molecules present on the adsorbent surface), while C_o is the initialconcentration of the solute.

3.5.4.3. Breunauer-emmet-teller (BET) model

A good number of adsorption models discussed in this work take account of adsorption with 'monolayer' coverage except for Freundlich isotherm and Dubinin-Radushkevich models.The BET theory is an extension of the Langmuir theory, which is single-layer molecular adsorption. One of the three assumptions used to develop the Langmuir model is adsorption only occurs at the unoccupied adsorption sites called a monolayer. The BET model was also developed using the same assumptions except for the single-layer adsorption coverage. The diagram below shows the multi-layer adsorption on a flat surface.

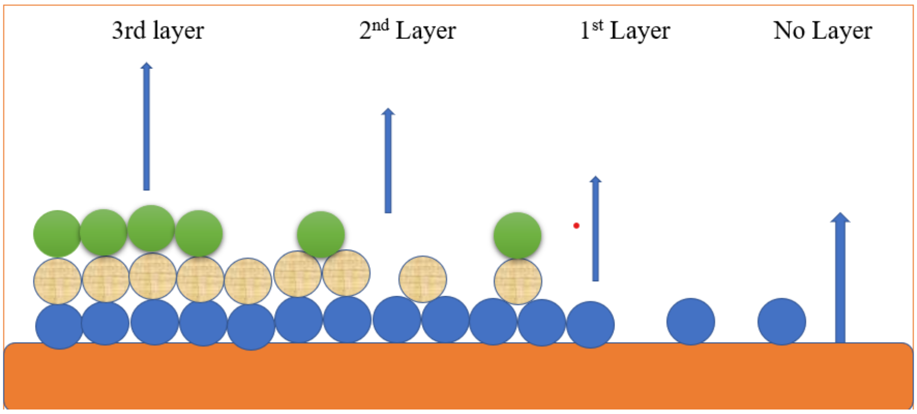

Figure 4. Modified multiple layering in BET theory (Do, 1998)

This theory was initially developed for adsorption in a gaseous phase, but it is now used in liquid-phase adsorption. In designing the BET model, the surface area of the flat surface is covered by no layer (S_o), one layer (S_1), two layers (S_2), three layers (S_3), and n layers of adsorbate molecules. This is then followed by applying the Langmuir model using the concept of the kinetic theory of adsorption and desorption to this multiple-layering process. The rate of adsorption on any layer is equal to the rate of desorption from that layer. The BET model for the liquid phase based on the above theory can be written as below:

$$\frac{q_e}{q_{max}} = \frac{BC_e}{(C_S^* - C_e)(1+(B-1)(C_e/C_S^*)} \quad (13)$$

where B and C_S^* are constants related to the energy of adsorption and the saturation concentration of solute, respectively.

If B and $B^*(C_e/C^*)$ are very high, the BET equation is simplified as:

$$\frac{q_e}{q_{max}} = \frac{1}{1 - C_e/C_S^*} \quad (14)$$

Many researchers are implementing this model to reciprocate the assimilation of many materials onto kaolinite clay. BET model has been used in research to point out the assimilation of phosphate from the aqueous solution using kaolinite composite (Gao et al., 2013). From the fitting of the experimental data, the values of parameter B were less than $1 (r^2 > 0.91)$. The parameter B relates to the energy of adsorption and can be written as:

$$B = exp\left(\frac{E_A - E_L}{RT}\right) \quad (15)$$

where E_A is the heat of adsorption in the first layer and E_L is the heat of liquefactionin subsequent layers.

3.5.4.4. Three-parameter isotherm equations

Sometimes the obtainable two parametric models are not enough to correlate the assimilation of experimental information, it is, therefore, essential to add or alter the available models. Many models with three or more parameters are now available. Some of them are Sips, Toth, Redlich-Peterson, Unilan, and Keller-Staudt. Two of these isotherm equations (Unilan and Keller-Staudt) have never been implemented to correlate the adsorption equilibria of hazardous substances in kaolinite composite.

3.5.4.4.1. Sips isotherm

Sip presented this equation in 1948 which he also named the Langmuir-Freunlich equation because it has characteristics that resemble Langmuir and Freundlich's isotherms. The Sips model makes up a better saturation function at an adequately high concentration.

$$q_e = q_{max} \frac{(K_s C_e)^{1/n_S}}{1+(K_s C_e)^{1/n_S}} \quad (16)$$

The parameter K_S represents the adsorption affinity, while the parameter n_S is regardedas the parameter characterizing the system's heterogeneity which might have resulted from the kaolinite mineral or solute, or a combination of both. If the system ishomogeneous ($n_S = 1$), the Sips equation above becomes Langmuir isotherm **(Eq.16)**.

The conducts or attributes of the Sips model at elevated and low solute concentrations are graphically represented in **figures 5 & 6** respectively.

Sips isotherm has been used by several researchers to interpret their adsorption experimental data(Bhattacharyya & Ray, 2015; Yao et al., 2014). Work has been done on the assimilation of methylene blue onto biochar-clay materials using kaolinite clay mineral and montmorillonite (Yao et al., 2014) in which the experimental data were adequately represented by this isotherm.

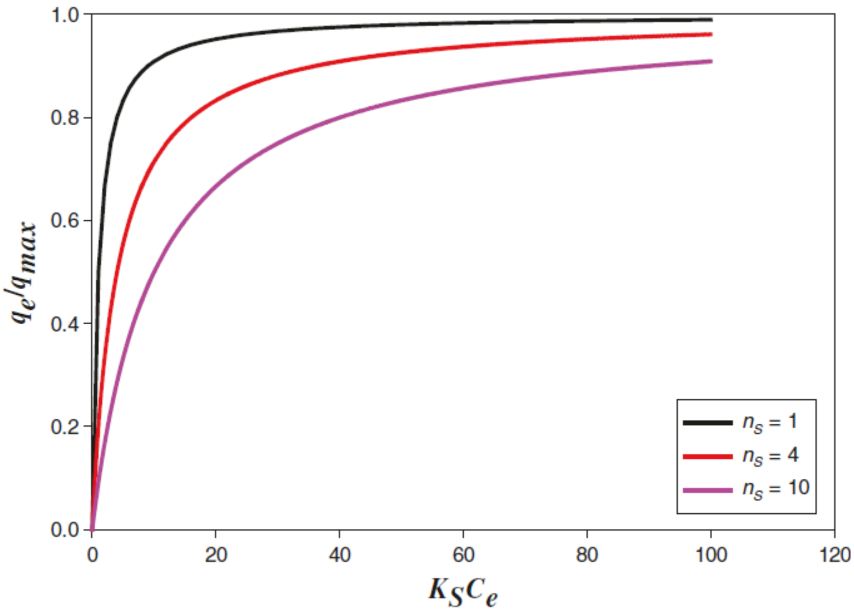

Figure 5. Graphical representation of Sips isotherm at elevated concentrations

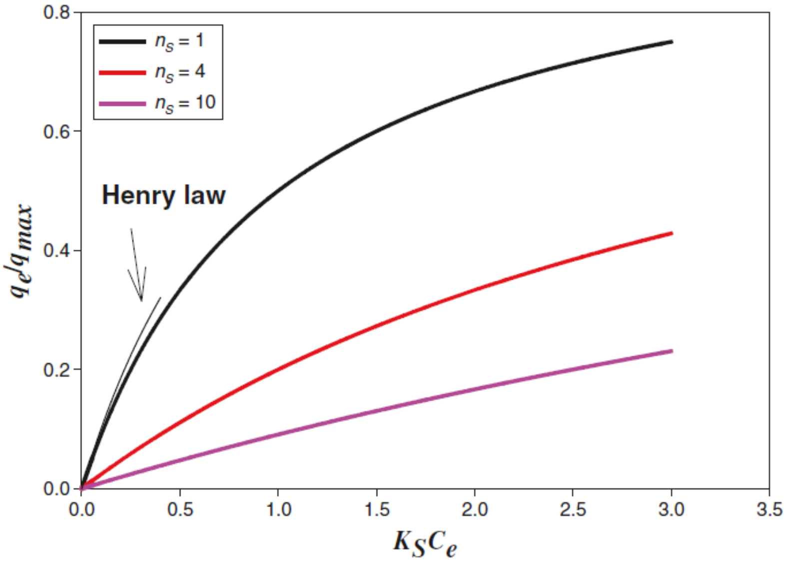

Figure 6. Graphical representation of Sips model at low concentrations

3.5.4.4.2. Toth model

Gaseous phase adsorption experimental data is usually interpreted by using the Toth equation. It is a three-parameter model that exactly depicts the attributes of Henry's law at low concentrations and narrow saturation performance at high concentrations. The mathematical expression of the Toth model is written as:

$$\frac{q_e}{q_{max}} = \frac{K_T C_e}{[1+(K_T C_e)^{nT}]^{1/nT}} \quad (17)$$

Parameter K_T is the adsorption affinity, and

n_T is a parameter representing the system heterogeneity.

These parameters are utilized for both adsorbate-adsorbent pairs where the parameter nT is less than one. The system would have been more heterogenous if the parameter deviated far from one. The Toth model reduces to the Langmuir isotherm if the parameter is equal to **1**. The Toth equation is a three-parameter model that is suitably used to explain a lot of liquid phase adsorption data of harmful substances (Chandra, Ju, Ayucitra, & Ismadji, 2013; Unuabonah & Adebowale, 2009).

3.5.4.4.3. Redlich peterson (r-p) isotherm

This is also a three-parameter equation that adequately represents adsorption equilibria over a wide range of concentrations. Its mathematical expression is written as:

$$q_e = \frac{K_R C_e}{1+a_R C_e^{\beta}} \qquad (18)$$

where K_R, a_R, and β are adsorption affinity, constant parameter, and parameter characterizing the heterogeneity of the system, respectively. Redlich-Peterson isotherm is a well-known model used to forecast the adsorption of harmful or hazardous materials onto kaolinite mineral composites (Kadu & Chikate, 2013; Peng et al., 2013). The Redlich-Peterson equation aggregates both the Langmuir and Freundlich characteristics to form one equation. It reduces to a linear isotherm at low surface coverage and to the Langmuir isotherm when β is equal to 1.

3.6 Adsorption process and characterization of clay minerals

The presence of minerals in kaolinite is nowadays simpler to identify and characterize due to the invention of new analytical equipment for scientific research purposes. There are many laboratory instruments employed to identify and quantify the presence of minerals in kaolinite clay. Some of these characterization instruments include nitrogen sorption, X-ray diffraction (XRD), scanning electron microscope (SEM), transmission electron microscope (TEM), infrared spectroscopy (IR), etc. In laboratory analysis, the pore volume, size, and surface area of a sample are ascertained by utilizing sorption isotherms of nitrogen gas at various pressure and temperature of 77.15 K of the nitrogen gas. A decrease in pressure reduces the amount of nitrogen gas in the system leading to what is referred to as desorption isotherm. There are various assimilation models normally employed to ascertain the pore sizes, structure, and surface area of samples in material science. The surface area of porous materials or adsorbents with an unknown surface character from the sorption of nitrogen gas is ascertained using two main methods which are BET and X theory methods. The BET equation is popularly used on the isotherms of nitrogen among others and has been in use since 1938. The formation of monolayer gas molecules on the solid surface is used to calculate the specific surface while the presence of pores, pore volume, and sizes are determined by using the principle of capillary distribution. BET models for the analysis of the surface area of porous materials are written as:

$$\frac{V}{Vm} = \frac{C\frac{P}{P_O}}{\left(1-\frac{P}{P_O}\right)\left[1+(C-1)\frac{P}{P_O}\right]} \qquad (19)$$

where
V is the volume of gas adsorbed at STP,
V_m is the volume of gas needed for monolayer adsorption,
P_o is the saturation pressure at the measured temperature,
P is adsorptive pressure, and C is a constant.

The linear form of equation (4.19)can be written as follows:

$$\frac{P/P_O}{V(1-\frac{P}{P_O})} = \frac{1}{CV_m} + \frac{C-1}{CV_m}(P/P_O) \quad (20)$$

The slope and intercept of Equation (4.20):

$$Slope_{BET} = \frac{C-1}{CV_m} \quad (21)$$

$$Int_{BET} = \frac{1}{CV_m} \quad (22)$$

V_m is accounted for by the expression below

$$V_m = \frac{1}{Slope_{BET} + Int_{BET}} \quad (23)$$

and

$$C = \frac{1}{V_m Int_{BET}} \quad (24)$$

For the BET surface area calculation, the value of V_m can be related to the number of moles of monolayer coverage. To convert the value of V_m into BET surface area the following equation is used:

$$S_{BET} = \frac{n_m a}{m} N_A = \frac{(V_m/22.400)a}{m} N_a \quad (25)$$

Where

S_{BET} is a BET surface area (m^2/g),

m is the mass of the sample (g),

N_A is the Avogadro number. The unit of V_m is cm^3/g. The effective molecular cross-sectional area, a is calculated by the following equation:

$$a = 1.091\left(\frac{M}{N_{AP}}\right) \quad (26)$$

Here, M and ρ are the molar mass of the gas and the liquid density of the gas, respectively, and the constant 1.091 is the packing factor. The application of this technique to determine the surface area of materials has existed for a very long time, but it is important to be careful with the data obtained to ensure accuracy (Michot & Villieras, 2006). The measured sorption isotherms of clay minerals also strongly depend on the operating conditions such as degassing temperature and time, the gas probe, and the type of clay minerals. The specific surface areas obtained by application of the BET theory to gas sorption isotherms on clay mineral systems are subject to significant variations in apparent molecular areas with variations in surface structure, exchangeable cation, and micro-porosity (Aylmore, 1974).

4. Search method

A sample of over 100 data articles related to this work was systematically reviewed. The samples were obtained from various online research database sources. The research question was formulated based on previous works that outline the "Utilization and Environmental Concerns of $Al_2Si_2O_5(OH)_4$ and its Adsorption Models in Toxic Environment".

5. Conclusion

Many works have been done on the use of kaolin in various applications ranging from ceramics, refractories, paper, cosmetics, white wares, pants, polymers, cement and concrete, agriculture, cosmetics, pharmaceuticals, etc. The global market and the demand for kaolin utilization are intense due to the driving forces such as paper industries and the expansion in urbanization and industrialization in developing countries. The expansion of the kaolin market size adds to the economic growth of kaolin-producing nations. According to a report from Fortune Business Insights, the kaolin market size is projected to rise to USD 5.7 billion in 2029. There are however a few health and environmental detriments connected with kaolin minerals from mining to utilization. Governmental bodies should adopt adequate measures to control the exposure of workers to particulate matter during kaolin mining. Exposure to dust will cause internal dust accumulations leading to severe health complications. Geologic and geochemical data is used to determine the potential detrimental problems from the use of kaolin clay minerals.

Kaolin is used as an absorbent to remove contaminants from wastewater and it also protects municipal landfills from the effects of chemicals from solid wastes. The theories of various equilibrium models such as Langmuir, Freundlich, etc, in the removal or adsorption of hazardous materials using kaolinite clay as an adsorbent, have been outlined but their experimental investigation was not covered because it is the scope and objectives of this dissertation. It is eaten by humans in some parts of the world to suppress hunger and as a decontaminant in some animal feed.

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