

Removal of Colorants Using Locally Activated Bentonite Clay: A Review

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Abstract

Bentonite clay is an aluminum phyllosilicate material that has been extensively studied as a suitable adsorbent for many pollutants. However, the activation of bentonite clay for color removal or bleaching has been reported to improve the porosity, surface area, and adsorption efficiencies. According to literature survey, physical, chemical and pillaring activations using diverse activating agents such as organic/inorganic acids, alkali, thermal or microwave heating, and so on have led to the preparation of highly improved bentonite clay composites, with superior adsorption capacities via cation exchange mechanism or physisorption/chemisorption mechanisms than commercially available adsorbent and inactivated bentonites. Furthermore, the activation of bentonites with biopolymers is an aspect of research that will develop new non-toxic, eco-friendly, and green adsorbent materials with enhanced tunable porosity, thermal stability, and adsorption capacities towards color removal in various oil extracts and gas condensates.

Keywords: Bentonite clay, Color removal, Adsorption, Bentonite activation

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I. Introduction

Gas condensate is a water-white or light straw-colored liquid that contains mostly hydrocarbon ranging from C₅-C₈ or higher and has an API gravity ranging from 50-75 degrees (Speight 2015). They are also called low-boiling naphtha or Ligroin that can be used as a feedstock for refinery upgrading processes as well as for the production of petrochemicals (Speight 2019). More importantly, color is a very critical feature that determines the quality, type, and marketability of petroleum or allied products i.e. products with poor color quality tend to be useless to both the producer and the customer (Bergeron *et al.* 1999). Therefore, it is imperative to remove associated foreign colorcontaminants in these products before delivery to the general public.

Adsorption is an economical method used in the petroleum industry and can be achieved at ambient temperature, pressure, and the use of cheap materials (Emam 2018). More so, bleaching has been used in the manufacturing industry for ages and has been beneficial for the removal of different impurities that make finished products unmarketable or regarded as low quality. The bleaching process is simply the removal of color or impurities from substances using natural material or chemical compounds. Numerous materials like carbons, silica gels, and alumina, which are usually amorphous in nature and containing a complex network of interconnected micropores, mesopores, and macropores have been extensively exploited as adsorbents. Nevertheless, zeolites with precise porosity and dimensions have been prepared in different forms as composite materials as suitable adsorbents.

Bentonite is a material containing aluminum phyllosilicate clay usually formed from the weathering of volcanic ash in the presence of water (**Figure 1**). Thus, bentonites are chiefly made of montmorillonite, which is regarded as major clay mineral of the dioctahedral smectites group consisting of a 2:1 layer clay formed by an octahedral (O) sheet comprising Al³⁺ and Mg²⁺ ions positioned amid two Si tetrahedral (T) sheets (Erdoğan Alver and Alver 2012). Bentonite has been effectively utilized in many fields such as insulating materials (Javed *et al.* 2020), bonding materials (Matei *et al.* 2019), plastics (Araújo *et al.* 2004), rubber catalyst (Ge *et al.* 2018), pharmaceuticals (Viseras and Lopez-Galindo 1999), cosmetics and skincare products (da Silva Favero *et al.* 2019; Moraes *et al.* 2017), waste treatment (Babu and Antony 2019; Gupt *et al.* 2020), electrochemical sensors (Vernekar *et al.* 2020) agriculture and forestry, and so on (Ptek *et al.* 2012; Shattar *et al.* 2020).

For the effective application of bentonite clays as adsorbent, they need to be subjected to an appropriate physical or chemical treatment otherwise known as activation to improve their surface properties. The application of activated bentonite in the manufacturing industry has been extensively investigated. In a review report, Hussin *et al.* (F. Hussin *et al.* 2011) highlighted the textural characteristics, surface chemistry, and activation of

bleaching earth towards the purification of vegetable oils. In the same vein, Akinwande *et al.* (Akinwande *et al.* 2015) showed that seed hulls (carbonaceous materials) can be used as an effective adsorbent either alone or in combination with other clay materials for the bleaching of vegetable oil. With more studies on the purification of vegetable oils and limited reports on fuel oil, (Emam 2018) revised extensively, the use of different clay materials as an adsorbent in different petroleum refining processes.

Need to say, natural clay material like bentonites is widespread across various parts of Nigeria has been under-utilized in processing plants and industries owing to limited research investigation in the area (Figure 2). Also, Nigeria has abundant gas condensate reserves that are not marketable yet due to its color and scarcity in the literature on the use of activated clay to remove color in petroleum products. Therefore, this review aims at reviewing various studies carried out on the removal of colorants in gas condensate and the extents of bleaching efficiency achieved using activated bentonite clay and also establish ranges of optimum conditions necessary to achieve a high color removal efficiency using activated bentonite clay.



Figure 1: Bentonitic clay chunks and bentonite powder

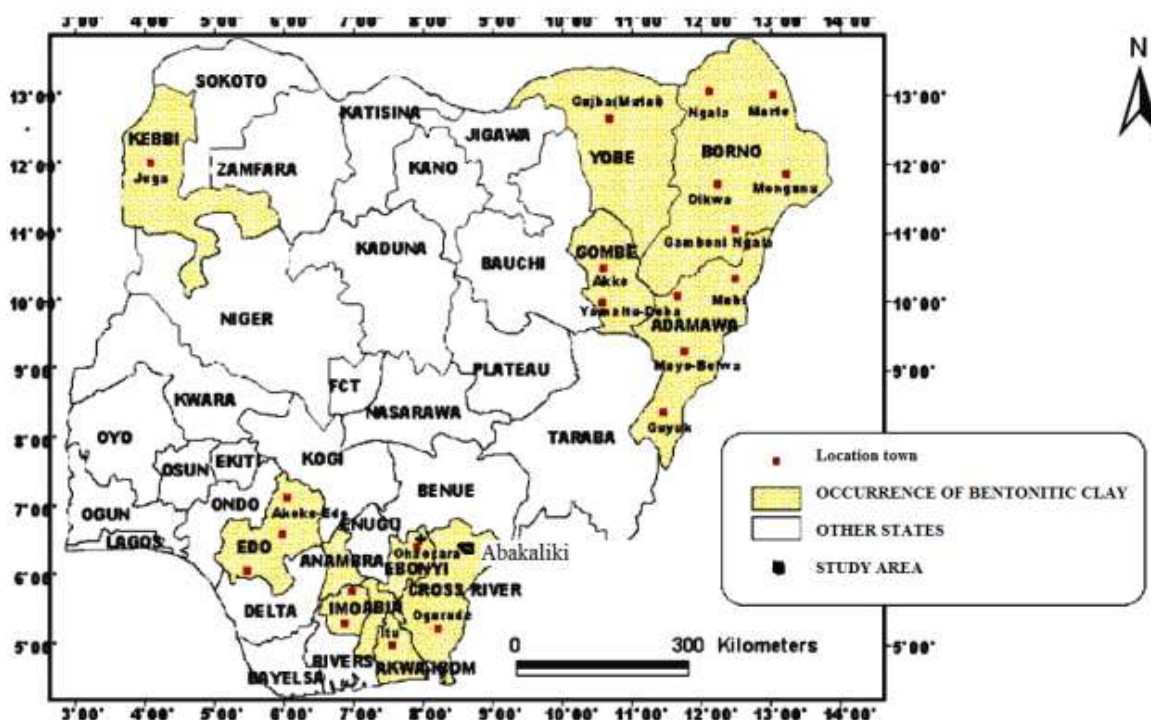


Figure 2: Map of Nigeria showing the distribution of bentonite clay. Reproduced from (Nweke *et al.* 2015). Copyright 2015 Creative Commons Attribution License 4.0 International License.

II. Bleaching And Bentonite Activation

The bleaching process of oil extracts (crude oil or edible oil) is aimed at absorbing all coloring components and to decompose hydro-peroxides compounds in the substance. This helps to remove contaminants such as oxidation products, phosphatides, pigments, metals, and soaps, etc. (Ma *et al.* 2017). Activation entails

the fine-tuning of the absorbing material to absorb color or impurities in oils and solutions through physical or chemical processes. Clays are minerals that contain alternating sheets of silica tetrahedral and aluminum octahedral, the silica tetrahedral is coordinated with oxygen atoms while the aluminum octahedral is coordinated with oxygen atoms or hydroxyls (Yildiz *et al.* 2004). Bentonite clays can be classified as sodium or calcium bentonite depending on the most dominant exchangeable ion present (Wu *et al.* 2006). The adsorptive capacity of activated bentonite clay can be affected by different properties like the type of clay, location of clay, impurities found in the clay, acid concentration, activation temperature, acid to clay ratio, time of activation, and surface area of the clay, etc. Also, the elemental composition of clay plays a major role in determining the type of clay, structural changes, and absorptive properties of the clay (Table 1). For example, the NaO/CaO ratio indicates the presence of calcium or sodium montmorillonite and the SiO₂/Al₂O₃ ratio indicates if the clay is suitable for bleaching, zeolite development, or drilling mud, etc.

The activation of bentonite is quite a complex phenomenon involving the substitution of exchangeable cations, Al³⁺, Mg²⁺, and Fe²⁺ against protons from the octahedral sheet, resulting in the improvement of bleaching effectiveness, greater hydrophobicity and porosity structure strongly protonated clay mineral surface, increase in specific surface area from the original 20–130 m²/g to higher than 200 m²/g, and generate higher affinity to the organic molecules (Figure 3) (Violeta *et al.* 2010). It is noteworthy that decolorization or bleaching process also involves catalytic reactions, ion exchange, and reactions due to surface acidity of the solid phase, thus it is not only a mere physical adsorption process (Christidis and Kosiari 2003). The activation of bentonite clay activation has been achieved by physical (thermal and microwave heating), chemical, and pillaring activation methods.

Table 1: Elemental Composition of Some Clays Minerals

Authors	Elemental Composition (%)										
	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	MgO	CaO	Na ₂ O	K ₂ O	TiO	MnO	LOI	CaO/Na ₂ O
Ajemba and Onukwuli (2012)	64.25	9.87	8.08	1.23	0.06	-	0.65	0.84	-	1.87	-
Caglar <i>et al.</i> (2013)	80.14	12.01	1.43	2.10	1.92	0.04	1.97	0.29	-	-	48
Motlagh <i>et al.</i> (2008)	67.8	11.48	2.18	2.50	1.67	2.76	0.19	-	-	-	0.61
Motlagh <i>et al.</i> (2011)	69.61	11.13	2.41	2.13	2.66	2.66	0.19	0.19	-	9.86	1
Meesuk and Vorasith (2006)	61.68	3.28	11.02	1.68	0.47	<0.10	1.64	10.05	0.12	5.59	0.47
	84.36	0.19	0.46	0.52	0.32	<0.10	0.37	6.39	<0.05	4.19	0.32
Mohammed <i>et al.</i> (2011)	56.77	15.67	5.12	3.42	4.48	1.11	0.60	-	-	9.49	4.04
Nwabanne and Ekwu (2013)	52.70	16.6	19.53	-	2.06	-	-	2.53	0.29	-	-
Usman <i>et al.</i> (2013)	41.47	36.92	0.07	0.48	0.60	0.05	0.08	-	-	20.33	12
Wu <i>et al.</i> (2006)	65.25	15.12	5.27	2.04	1.67	2.71	2.07	0.68	0.21	4.92	0.62

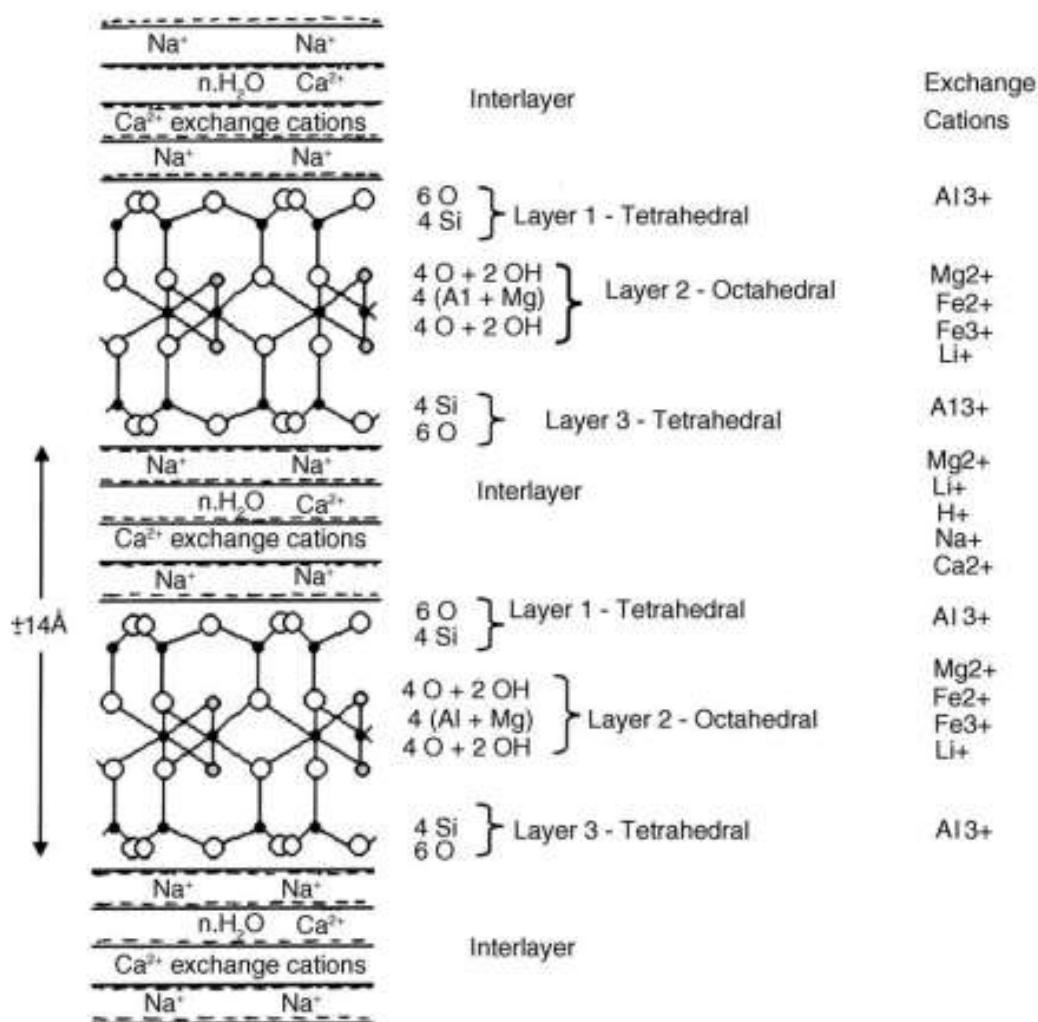


Figure 3: Structure of montmorillonite clay. Reproduced from (Tyagi *et al.* 2006). Copyrights 2005 Elsevier B.V.

2.1 Physical Activation

This is the heating of clays at a particular range of temperatures (200-1000°C) for about 2-4 h causing the clay to undergo dihydroxylation. During thermal activation, structural changes are observed in the clay mineral depending on certain factors which include but are not limited to heating rate, cooling rate, holding temperature, and time (Khalifa *et al.* 2020). Physical activation of clays has been achieved to a large extent by either thermal or microwave heating.

In a study, Sarikaya *et al.* (Sarikaya *et al.* 2000) identified that the heating of white calcium bentonite from the Kijtahya, Turkey, at various temperatures between 100-1300°C for 2hr causes deformation (i.e. changes of the clay by dehydration, dehydroxylation, shrinkage, and fracture, etc.) of the crystal structure of the montmorillonite-content depending on temperature (up to 700 °C). Also, Bayramet *al.* (Bayramet *al.* 2010) reported that during the thermal analysis of calcium bentonite, dehydroxylation occurred between 300-750°C and a complete collapse of the 2:1 smectite at 900°C, thereby affecting the crystal structure of the clay. Similar results were obtained by Önal and Sarikaya 2007 (Önal and Sarikaya 2007) during the thermal activation of Turkish bentonite. They reported that at 400 °C dehydration of the smectite structure was reversible but at 600 °C the changes became irreversibly and finally a total collapse of the structure at 900°C. Bello *et al.* (Bello *et al.* 2011) recorded a color reduction in cottonseed oil using thermally activated clay at 145 °C which gave an increase of bleaching efficiency from 14.9% to 49.6%; while Taher *et al.*, 2018 (Taher *et al.* 2018) also thermally activated Indonesian bentonite for adsorption of procion red from an aqueous solution at a temperature of 500°C. The results showed that there was no significant change in the structure of the raw bentonite and activated bentonite, however, adsorption using the activated bentonite was highly efficient at different pH (**Figure 4**). Meanwhile, Masindi and Ramakokovhu (Masindi and Ramakokovhu 2020) noted that thermally

activated and vibratory ball milled South African bentonite clay recorded > 99% adsorption efficiency which reached 62.5 mg/g and is superior to other bentonite families. Another study by Olu-Owolabi *et al.* (Olu-Owolabi *et al.* 2016) shows that the calcination of bentonite clay modified with *Carica papaya* seeds or pine cones produced new adsorbents with reduced surface area that were not significantly affected by pH between 3 and 7. Also, undesired properties such as adsorbent bleeding were removed and mechanical strength was enhanced.

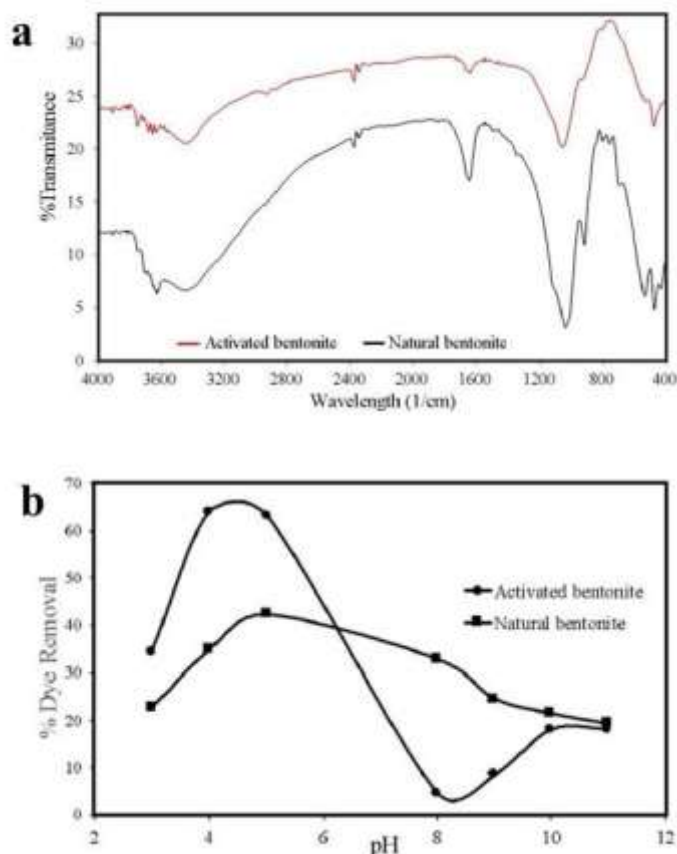


Figure 4: (a) FT-IR Spectra of (b) Effect of initial pH on the adsorption of procion red onto natural bentonite (NB) and thermal activated bentonite (AB). Reproduced from (Taher *et al.* 2018). Copyrights 2018 Creative Commons Attribution-Non-Commercial 4.0.

Nevertheless, microwave energy (0.3 MHz to 300 GHz) has been developed as a new tool for high-temperature processing of materials with a lot of advantages such as rapid and uniform heating; decreased sintering temperatures; improved physical and mechanical properties (Clark *et al.* 2000). Baldassari *et al.* (Baldassari *et al.* 2006) affirm that microwave heating (MWH) of organoclays proffers more improved results during the treatment of higher-charged clays, however, no significant differences in terms of kinetics of intercalation with lower-charged clays were observed when they compared the preparation of organoclays using both conventional–hydrothermal and microwave hydrothermal processes. Also, Zemanová *et al.* (Zemanová *et al.* 2006) found out that the MWH (2.45 GHz) dispersions of Li-saturated montmorillonites showed a slightly higher layer charge reduction than conventionally heated samples with no significant change observed in the layer charge. Similarly, a series of solid powders with different interlayer lithium content treated with microwaves (30.0 GHz) revealed high layer charge reduction than conventionally heated powders as the efficiency was enhanced by increasing interlayer lithium content (Zemanová *et al.* 2006). In another study, Yapar (Yapar 2009) prepared organoclays based on hexadecyltrimethylammoniumbromide, smectite from central Anatolia, and microwave irradiation at 360 W for 5 min. The physicochemical properties of the obtained clay reveal *d*-spacings of 19.5 Å and ion exchange efficiency up to 70% which is comparable to the organic cation exchange efficiencies reported using conventional techniques. Furthermore, Korichi *et al.* (Korichi *et al.* 2009) characterized smectite after activating with 2M HCl several times under varied MWH power. It was reported that the obtained material had microporous and mesoporous structures with specific surface area, pore diameter, and pore volume of a suitable adsorbent given as 165.80 m²/g, 50 nm, and 0.1919 cm³/g, respectively,

while the pore volumes of mesopores and micropores were 163.90×10^{-3} and 28.89×10^{-3} cm^3/g , respectively. Additionally, Gunawan *et al.* (Gunawan *et al.* 2010) asserted that organo-bentonites obtained from MWH are more time-efficient than those of normal heating. However, they further observed that the modification of bentonite with linear alkylbenzenesulfonate (LAS) was more effective as a bleaching agent than cetyltrimethylammonium bromide (CTAB) used for the **modification of Bentonite Pacitan**.

2.2 Chemical Activation

This is the treatment of material with chemical activating agents such as mineral acids and then washing with water to remove excess acid/alkali from the clay. The clay is then dried and sieved again to obtain a uniform particle size. Chemical activation also involves the modification of surface functional groups and structure (Ademiluyi and David-West 2012).

2.2.1 Acid Activation

This is the chemical modification of clays with a mineral acid with the aim of increasing porosity; surface acidity. Acid activation of bentonites is the most extensively studied and one of the most important processes that have been performed to achieve desirable structural manipulation of bentonite clay materials for industrial applications and adsorption studies (Ptek *et al.* 2012). In the advent of acid treatment of the clay materials, the interlayer cations are being replaced with H^+ ions followed by partial destruction or de-lamination of the aluminum octahedral and silicon tetrahedral sheets with the subsequent dissolution of structural cations, thereby improving the adsorption properties and capacity towards color-producing substances from the oil extract (Amari *et al.* 2018; Foletto *et al.* 2011; Motlagh *et al.* 2008). In an investigation, Caglar *et al.* (Caglar *et al.* 2013) observed that after the acid activation of Uney bentonite, a considerable amount of Al was retained as a constituent in acid-resistant impurities following the decomposition of the montmorillonite. It was recorded that mesoporous impurities with a maximum surface area of 303.9 ± 0.4 $\text{m}^2 \text{g}^{-1}$ were observed which caused a sharp decrease in the d_{001} lattice spacing of the montmorillonite to 15.33 Å and a reduction of the crystallinity in the activated products. In another study, Ajemba and Onukwuli (R O Ajemba and Onukwuli 2012) studied the adsorptive performance of Nteje clay and opined that acid treatment involves two steps namely; a) Dissolution of the Al_2O_3 , CaO, MgO, and other ions. b) Gradual exchange of the Ca, Al, Mg ions with hydrogen ions from the mineral acid. Thus, during acid activation, chemical changes cause spaces in the crystal lattice which increases the adsorptive power of the activated bentonite (Taha *et al.* 2011). Further studies by Ajemba and Onukwuli (Regina O. Ajemba and Onukwuli 2014) reveals that acid-leaching with HCl triggered several modifications of the clay from Udi, Nigeria, and increased the surface area, Si/[Al + Fe + Mg] ratio, and adsorption capacity from 29.8% to 66.7%. Similarly, the acid-leached sample prepared with 7M HCl yielded an adsorbent that was even more efficient in bleaching palm oil under the experimental conditions.

Fernandes *et al.* (J. V. Fernandes *et al.* 2020) demonstrated that the acid treatment of natural calcium bentonite with HCl or H_2SO_4 was efficient for increasing the adsorption capacity of anionic dye but the acid functionalization of bentonite did not significantly modify the adsorption of cationic dye. Also, (Tayebee and Mazruy 2018) recorded that acid and thermal activation of natural nano-bentonite increases the surface area >220 m^2/g for the spontaneous and effective adsorption of colored dye via an endothermic process. Khan (Khan 2015) activated Pakistan clay using four organic acids namely oxalic, phosphoric, acetic, and citric acid at 1N for 0.5h. It was reported that the oxalic acid gave the best adsorption capacity compared to the acids used in the bleaching of the vegetable oil. Wu *et al.* (Wu *et al.* 2006) from their study of the characterization, acid activation, and bleaching performance of Xinjiang Bentonite reported that the NaO/CaO ratio of the bentonite clay was greater than 1 indicating the presence of swelling bentonite. The bentonite clay was found to contain 56.7% montmorillonite which was mainly sodium characterizing it as sodium bentonite with an optimum bleaching capacity achieved at 83.2%. Nwabunne *et al.* (Nwabanne and Ekwu 2013) investigated the bleaching of palm oil using Nteje clay activated with 1M H_2SO_4 and reported that temperature, activation time, and dosage increased the bleaching efficiency while the increase in particle size led to a percentage decrease. Further, Nguetnkam *et al.* (Nguetnkam *et al.* 2008) decolorized palm oil, cottonseed oil, and maize germ oil at different temperatures, acid concentrations, and times. They concluded that the palm oil, cottonseed oil, and maize germ oil were completely bleached at a temperature between 85°C – 95°C within 0.25 – 2.5h giving a bleaching efficiency of 80%; while Li *et al.* (J. Li *et al.* 2008) in their research on the decoloration of biopetroleum using acid-activated bentonite reported that the bentonite was effective in removing color-induced components such as polycyclic aromatic hydrocarbon, quinone, unsaturated carbonyl groups, and so many others. This gave a maximum color efficiency of 83.31% and it was concluded that the increased color removal percentage was due to the surface area and acidity of the clay after activation.

Additionally, Motlagh and colleagues studied the characterization and bleaching efficiency of Iranian bentonite clay using different spectroscopic techniques. They discovered that the increase in HCl concentration up to 5N showed an increase in octahedral cations dissolution which increased the percentage of color adsorbed

up to 88.9%. They also activated colza-soybean oil with H₂SO₄ and reported that at 7N, the octahedral sheet was affected by acid activation resulting in the dissolution of cations where the Mg²⁺ was more affected than the Al³⁺ and Fe³⁺ ions. (Motlagh *et al.* 2011; Motlagh *et al.* 2008). Also, Foletto *et al.* (Foletto *et al.* 2011) studied the adsorption of sunflower oil using activated acid bentonite. They compared two bentonite samples and the experimental findings showed that clays with higher MgO content are more activated by mineral acids than those of smectites with lower MgO. They concluded that the structural change and bleaching efficiency of the bentonite was dependent on the acid concentration. They also studied activated bentonite from Mendoza Argentine using H₂SO₄ (4N, 8N) at 2 and 3.5h and observed that the optimum bleaching efficiency was 88% at 3.5 h using 8NH₂SO₄. This showed that destruction of the bentonite and bleaching efficiency was dependent on acid concentration and activation time. **Table 2** summarizes the reported optimum condition and amount of color absorbed according to some researchers.

Table 2: Some reported optimum conditions for bentonite activation

Authors	Type of Acid/alkali	Concentration (M)	Optimum acid conc.	% Color adsorbed
Ajemba and Onukwuli (2012)	HCl	1-5	3	88.9
Ajemba and Onukwuli (2014)	HCl	1-7	7	66.7
Bello <i>et al.</i> (2011)	Inorganic HCl	1.4 - 8.2	1.4	84.5
Foletto <i>et al.</i> (2003, 2011)	Inorganic H ₂ SO ₄	4,8	8	48.68
James <i>et al.</i> (2008)	H ₂ SO ₄	2 - 11	5	96
Motlagh <i>et al.</i> (2008)	Inorganic HCl	2-7	5	88.9
Motlagh <i>et al.</i> (2011)	Inorganic H ₂ SO ₄	2-7	7	102.6
Mohammed <i>et al.</i> (2011)	Inorganic H ₂ SO ₄	0.51 - 7.14	1.02	70
Nguetnkam <i>et al.</i> (2008)	H ₂ SO ₄	0.5 - 4	0.25, 0.50, 2.5	80
Okwara and Osoka (2006)	NaOH	1- 4	1	78.66
Salawudeen <i>et al.</i> (2007)	Inorganic H ₂ SO ₄ HCl	1.53 - 4.59 4.10 - 12.30	3.57	94.20 89.46
Usman <i>et al.</i> (2012)	HCl	0.5 - 3	3	27.27
Wu <i>et al.</i> (2006)	Inorganic H ₂ SO ₄	1.53 - 4.59	2.55	83.2

2.2.2 Alkaline Activation

Alkaline activation is the treatment of bentonite clay materials with basic solutions. Though not many reports on the alkaline activation on bentonite clays are available, this approach is also suitable for bleaching oil extracts. Gates and Bouazza (Gates and Bouazza 2010) argued that strong alkaline solution pH causes changes to the mineralogy of bentonites which might impact their performance as environmental barriers. However, the reaction between bentonite and alkaline leachates often results in pore filling and dissolution formation of hydrous hydroxy-aluminosilicate and hydrous carbonate mineral phases. Meanwhile, Anh *et al.* (Anh *et al.* 2017) discovered that alkaline solutions (pH = 8.4–13.1) cause almost no change in the montmorillonite fraction of the bentonite irrespective of the type of solution. More so, Aldabsheh *et al.* (Aldabsheh *et al.* 2015) observed the dissolution behavior of three Jordanian clay-rich raw materials in different alkaline solutions of 2, 5, and 10 M NaOH and KOH at 25 °C. The investigations reveal that the clay materials were to some degree soluble in concentrated alkaline solutions and a higher extent of dissolution was observed with increasing time and temperature. However, the extent of dissolution was somewhat higher in NaOH than in KOH solution. In addition, Bauer and Berger (Bauer and Berger 1998) also confirmed the dissolution of smectite clay in high alkaline concentration over kaolin in batch reactors operated at 35 and 80°C.

In a study, Okwara and Osoka (Okwara and Osoka 2006) bleached palm oil using alkali-activated kaolin, fuller's earth, and bentonite. They reported that at 3M, 2.9M, and 1M NaOH the kaolin, fuller's earth, and bentonite gave an optimum color removal of 76.38, 80.69, and 78.66 respectively. Novikova and Korsakov 2003 (Novikova and Korsakov 2003) examined the influence of activation on the structural and functional composition of Cambrian clay by treatment with sulfuric acid and sodium hydroxide solutions. The obtained results show that the crystallinity of the clay after acid-base treatment was unhindered according to x-ray diffraction studies. However, the content of the active centers and specific surface area decrease appreciably after acid treatment; while the base treatment increases the specific surface area and the content of the Lewis acid and base centers.

2.3 Pillaring Activation

Pillaring is a type of clay modification technique that employs both physical and chemical activation procedures to improve the properties of bleaching earth. This process improves the physicochemical properties, porosity as well as retention of the layer structure to enhance the capacity of the clay material to encourage adsorption of specific ions (C. Fernandes *et al.* 2007).

The mechanism of pillaring activation entails cation exchange and stabilization. The process involves inserting polycations into the basal space by a cation-exchange mechanism or by forming polycations *in situ* at the cation exchange capacity sites of the layered clay structure, after which the hydrated polycation pillars can be stabilized by high temperature (>400°C) heat treatment (Occelli *et al.* 2000). Clays pillared with metal oxides are of great importance because of their high thermal stability, high surface area, and intrinsic catalytic and adsorptive properties. These materials are made of intercalated species that are capable of preventing the collapse of the interlayer spaces, propping open the layers as pillars, and forming interlayer space (Bhattacharyya and Gupta 2008). Depending on the pillar species used, the resultant pillared clay has interlayer free spacings that are expanded to 4-20 Å and surface areas of 200-400 m²/g. The pore size of pillared clay depends on the calcination temperature; at high calcination temperature (>700°C) the microporous structure collapses and results in large pores, whereas pillared clays calcined at 400-600 °C behave as bi-dimensional molecular sieves (Romero *et al.* 2006; Xu and Zeng 2000).

For example, Falaras *et al.* (Falaras *et al.* 2000) reported that Al-pillared clay treated with sulphuric acid solutions provided improved bleaching efficiency for cottonseed oil better than untreated clay and commercially available Tonsil Optimum 214 clay. Similarly, Caglayan *et al.* (Caglayan *et al.* 2005) reported that the optimal pillaring process conditions for cottonseed bleaching were an initial concentration of AlCl₃ of 0.5 M, an OH⁻/Al³⁺ molar ratio of 0.3, and a thermal treatment temperature of 700°C. At these conditions, the bleaching efficiency of bentonite increased from 11.8 to 17.5% by acid activation and 33.5% by further Al pillaring.

Furthermore, Ding *et al.* (Ding *et al.* 2015) employed a one-step high-temperature and high-pressure method to synthesize AlCr composite pillaring agents by ion exchange. This approach which greatly reduced the steps and material consumption in conventional approaches also produced a novel material with improved adsorption properties. In a new syntheses and economical approach, Bertella and Pergher (Bertella and Pergher 2017) observed that a pillaring solution can be reused after filtration and precipitation with Na₂SO₄ solution even after being stored for up to one year will produce Al-pillared clays with basal spacings larger than 17 Å and BET surface areas greater than 160 m²/g. In the revised work of Klopogge *et al.* (Klopogge *et al.* 2005), they hypothesized that the modification of clay minerals by propping apart the clay layers with an inorganic complex gives rise to two-dimensional porous structured materials with acidic properties comparable to that of zeolites and is suitable for the cracking of vegetable oils for transesterification into biodiesel.

2.4 Biopolymeric Activation

Biopolymeric activation of bentonite clay to form a new class of hybrid composites is a novel technique that can be used for the adsorption of color in gas condensate and other oil extracts. These biopolymeric compounds can reverse the toxicity challenges provided by acid/alkaline activation by providing biodegradability, eco-friendliness, and non-toxic hybrid composites. The activation of bentonite with polymers via encapsulation improves the adsorption capacity and thermal stability of the new hybrid composite (Pandi and Viswanathan 2014; Tezcan *et al.* 2012). For example, alginate is a polymer with excellent gelation properties suitable for adsorption, thus, Aichour and Zaghouane-Boudiaf (Aichour and Zaghouane-Boudiaf 2020) reported that bentonite/alginate composite improved the elimination of dye stuff with high regeneration efficiency. In the same vein, Fabryanty *et al.* (Fabryanty *et al.* 2017) argued that microwave irradiation aids the effective formation of sorbent pores for color permeation in bentonite/alginate composite matrix. Their study shows that the variation in the mass of bentonite and temperature of irradiation affects the structural porosity of nanocomposites, and also increases the adsorption ability (**Figure 5**). According to Tan and Ting (Tan and Ting 2014), the immobilization of alginate onto bentonite improves sorption ability compared with the individual alginate beads within 6 h at pH between 4 and 5. More so, Lezehari *et al.* (Lezehari *et al.* 2010) confirmed that the surface modification of biopolymers by encapsulating with pillared clays proffers an enhanced adsorption capacity after cross-linking sodium alginate, aluminum pillared clay (Al-MontPILC) or surfactant-modified pillared clay (CTAB-Al-Mont-PILC) and divalent cation (Ca²⁺) to fabricate gel-like beads (**Figure 6**). Also, the impregnation of silk fibroin into bentonite produces plate aggregates to rosette-like arranged composites with intriguing monolayer adsorption capacity than either of the materials alone (Wahab *et al.* 2019). In a recent study, da Silva *et al.* (da Silva *et al.* 2021) demonstrated that the intercalation of bentonite with different proportions of chitosan via microwave heating leads to the development of bionanocomposite with good adsorptive properties aided by cation exchange. Also, by employing plasma-induced methods, Yang *et al.* (Yang *et al.* 2016) successfully grafted chitosan onto magnetic bentonite. The novel composite with intriguing magnetic properties, low turbidity, and high stability in an aqueous solution also displayed substantial adsorption properties for remediation studies. Besides, Li *et al.* (W. Li *et al.* 2018) designed gelatin/bentonite composite beads with high thermal stability and enhanced adsorption efficiency towards the removal of congo red (CR) and methylene blue (MB) at varied pH and contact time (**Figure 7**).

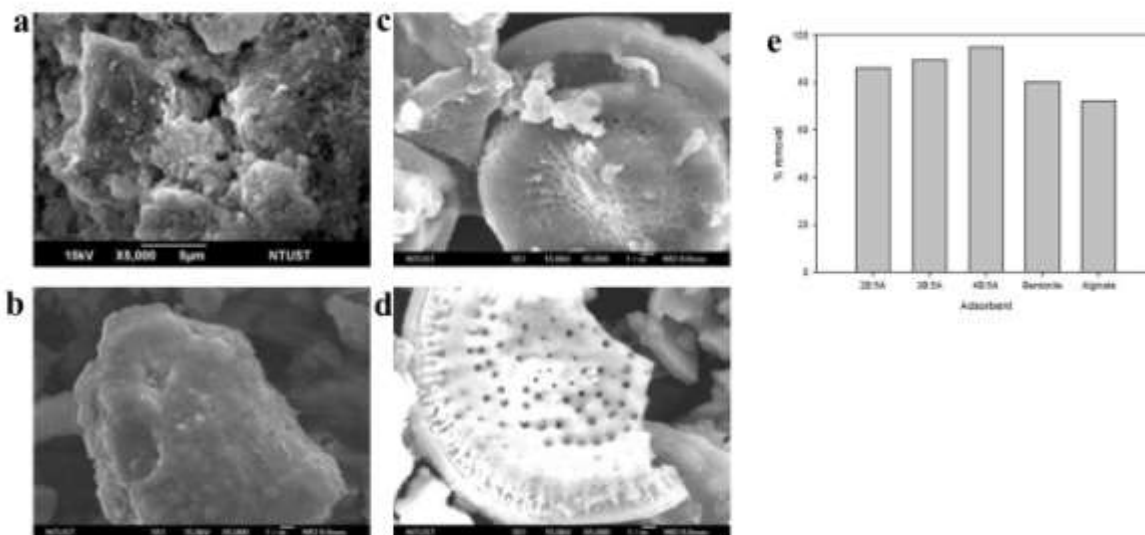


Figure 5:SEM images of (a) bentonite. Bentonite clay: sodium alginate ratios at (b) 4B:5A composite (c) 3B:5A composite (d)2B:5A composite (e) Removal of crystal violet from simulated wastewater. Reproduced from (Fabryanty *et al.* 2017). Copyright 2017 Elsevier Ltd.

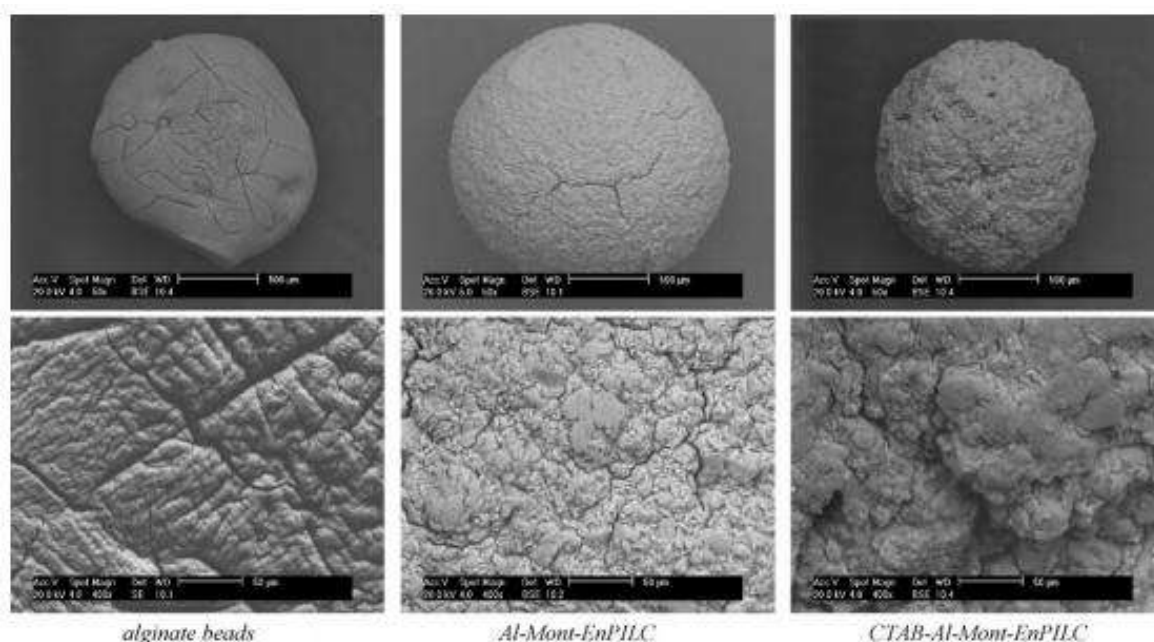


Figure 6:SEM images of alginate microbeads, encapsulated aluminium pillared clay (Al-Mont-EnPILC), and surfactant-modified pillared clay (CTAB-Al-Mont-EnPILC). 50x and400x magnification. Reproduced from (Lezehari *et al.* 2010). Copyright 2010 Elsevier B.V.

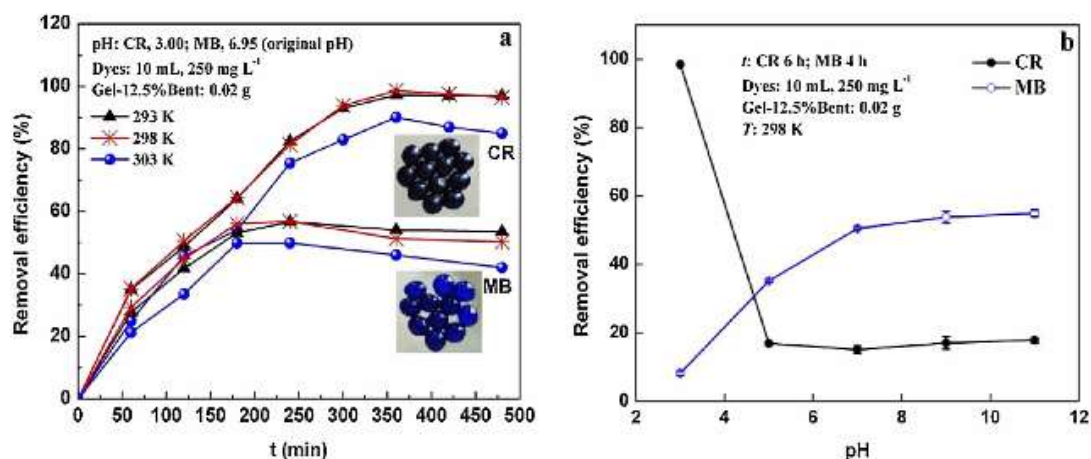


Figure 7: Investigations on the effects of (a) contact time and temperature (b), on the color removal efficiency of CR and MB. Reproduced from (W. Li *et al.* 2018). Copyrights 2018 Elsevier B.V.

III. Parameters Affecting Bleaching Efficiency

A lot of parameters are considered in designing the optimum reaction conditions for any bleaching process. The concentration of the acid or alkali used in activating plays a major role in affecting the bleaching efficiency of any bentonite clay. Aliu *et al.* (Aliu *et al.* 2016) opined that varying pH increases the acidity and increases the active surface sites. Thus, the rise of the pH continuously increases the cation exchange capacity of bentonite, thereby increasing adsorption. Nevertheless, Usman *et al.* (Usman *et al.* 2012) considered the potentials of using clay characterization as a major tool to predict its suitability for bleaching of vegetable oils. Although the spectroscopic characterizations revealed the composition of the clay, the conducted laboratory tests for bleaching performance confirmed the poor bleaching ability of the natural clay due to a low color reduction of 9.1%. However, activation of the clay with 3M HCl increases the decolouration to 27.3% a value still very low for effective bleaching.

Amongst other parameters, the type of acid, treatment time, treatment temperature, solid-liquid ratio, and moisture content is a crucial parameter that determines the optimum conditions for color removal (F. B. Hussin 2013). In a study, Eloussaief *et al.* (Eloussaief *et al.* 2020) regulated certain parameters such as HCl concentration (1–5 N), acid contact time (1–6 h), and temperature (40–90 °C) to obtain the maximum specific surface area of the activated clay. It was reported that 87% decolourization was obtained with the clay treated with 3 N HCl for 4 h at 70 °C. Also, Permana *et al.* (Permana *et al.* 2018) studied the adsorption of β -carotene from palm oil onto bentonite by monitoring the bleaching conditions like concentrations of β -carotene in palm oil (300, 400, 500 mg/kg), the dosage of bentonite (0.5, 1.25, 2 %), surface areas of bentonite (190, 225, 275 m²/g), and temperatures (50, 80, 110 °C). Under these conditions, the authors observed that an intraparticle diffusion model best describes the adsorption process with a low adsorption rate and a low average value of the coefficient of determination.

Furthermore, Mustapha *et al.* (Mustapha *et al.* 2013) evaluated the potentials of varying strengths (20, 30, and 45%) of HCl and H₂SO₄ acids towards the activation of clay and diatomaceous earth samples. The authors recorded a color reduction of 96.5 and 98% in a two-step adsorptive bleaching sequence with HCl. Similarly, Salawudeen *et al.* (Salawudeen *et al.* 2007) observed that HCl is a better clay activating agent than H₂SO₄, giving a 94.28% decolouration at 45% acid concentration after monitoring the effect of acid type, acid concentration, and mass of adsorbent towards palm oil bleaching. Meanwhile, Tebandeke and Tebandeke (Tebandeke IZ and Tebandeke H 2016) varied the strengths of 0, 10, 20, and 30% HCl and H₂SO₄ acids for the activation of local Ugandan clay from Kangole, towards the bleaching of cotton and sunflower seed oils. The study unveiled a maximum decrease in absorbance of bleached oil attained with clay leached in 30% sulfuric acid when the oil was in contact with clay for 30 to 40 min at 90 °C.

More so, in the studies of Meesuk and Vorasith (Meesuk and Vorasith 2006), the optimum bentonite refluxing of 5 hrs with sulfuric acid for activation to yield a bentonite suspension with a pH value of 3 was able to adsorb impurities and toxic compounds in repeatedly used palm oil at 80–85 °C within 30 min. In a comparative study, Usman *et al.* (Usman *et al.* 2013) explored the effects of adsorbent dosage (2 – 8 %), contact time (0 – 1.5 hrs), and temperature (60 – 90 °C) on pigment adsorption using Afashio kaolin (AFK) and local bentonite (BN). The results show that the optimum bleaching condition gave an efficiency of 81.4% and 62.4% for the BN and AFK clay respectively.

IV. Conclusion And Outlook

This review has considered the adsorption of color as a contaminant in oil extracts using bentonite clay. It has shown the different activation of bentonite clays to improve its physicochemical and adsorption efficiency. An extensive literature survey has shown that thermal and acid treatment of the adsorbent has been explored by many researchers. More so, the performance efficiency of the treated and untreated bentonite largely depends on different experimental conditions such as pH, temperature, porosity, etc. This review has also postulated that the toxicity issues attributed to organic and inorganic acids and alkali used for bentonite activation or treatment can be turned by the urgent utilization of cheap, readily and locally available, biodegradable biopolymer. Furthermore, bentonite clay can be activated/treated with biopolymers via encapsulation. This will open new avenues towards the fabrication of novel composite materials with enhanced adsorption efficiency and thermal stability. These low-cost modified bentonite clay adsorbents will offer numerous positive benefits that can be employed commercially for industrial purposes. Additionally, based on the arrays of existing research on the bleaching of oil extracts from different parts of the world, there are scanty reports on the bleaching of gas condensate which is a hot spot for geologists, especially in Nigeria. Therefore, this is a wake-up call for the urgent utilization of Nigeria's wide distribution of bentonite clay for the purpose.

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