

# THE CHEMISTRY OF VIGAN CLAY

*Consoladora P. Ridulme*  
*Praxedes Peralta*

## INTRODUCTION

The Philippines is endowed with abundant natural resources, most of which are untapped, unutilized, or even unexplored. Discovery of these mineral wealth and their proper use employing modern techniques in the production of serviceable goods would make the country one of the newly-industrialized countries in Asia.

Clay is one of the natural resources which is indigenous in the Ilocos Region, but which is not utilized to the maximum. This is a naturally occurring material composed of very fine particles, which are mainly clay minerals.

The craft of making clay pots was introduced in the Philippines as early as 1500 BC **when** Pre-hispanic Filipinos made drinking cups, jugs for storing water and grains, burial jars and **cooking pots**. Today, these remaining artifacts **are** the precious treasures found in museums.

The old Spanish houses of which Vigan is well known for and the many Catholic churches in the Ilocos Region that are made of bricks and clay tiles are evidences of the abundance of clay deposits in this region which **were** used in the construction of these edifices. These buildings which are several centuries old, have continuously withstood the elements of time and weather as well as man-made disasters. This proves the durability of clay as architectural materials. Maximum use of clay in this region with the application of modern technology in the production of clay products, particularly in the architectural industry, would lower the cost of building constructions. This would also solve the housing **problem** caused by the log ban program of the **government**

Many families in Vigan who are residing in Barangays Pagburnayan, Salindag and Bulala are engaged in making clay products like cooking wares, dinnerwares, pipes and other articles. However, traditional techniques are still applied in making these articles due to inadequate knowledge of the rural folks on the basic concepts of clay properties as well as the lack of technological skills and facilities. An in-depth training of these individuals on modern technological skills on the use of clay materials **and** the availability of modern facilities would lead to the production of clay products that are of higher quality.

Many clay deposits in the Ilocos Region are not utilized. In a research utilization seminar conducted by the UNP Research Center in the interior towns of Ilocos Sur and even in Abra, abundance of clay deposits was noted on the mountains and hillsides of these localities. Some people in these areas are aware of the presence of these minerals but they lack the knowledge and skills in using them as raw materials in making clay articles.

Experimental investigation of samples of these minerals as to their chemical properties and their proper uses would generate employment and increase the income of the farmers.

Hence, one of the proposed projects **of** the UNP Research Center is to undertake an experimental study on the properties; varieties and proper uses of clay deposits in this region and to launch the transfer of new technology relative to production of clay articles in the form of Research Utilization Seminar in the municipalities where clay deposits are abundant. This will give the people in these municipalities basic knowledge on the manufac-

ture of clay products, particularly ceramic and construction materials like red bricks, terra cotta, vases, tiles and pipes.

In connection with these activities, this paper expounds on the nature and composition of clay deposits, their chemical transformation and chemical structure and properties. The different types of clay mineral groups and the common varieties and uses of clay materials are described. Some chemical properties of clay samples taken from two barangays in Vigan, Ilocos Sur are also presented in this paper.

## NATURE AND COMPOSITION OF CLAY

Clay is a material of widespread occurrence. It is derived from the earth's surface, as a part of the lithosphere. It originates from rocks, rich in aluminum and silicon by the process of mechanical and chemical weathering. It is an active mineral portion of soil.

When rocks are broken or crumbled by heavy rain falling over a long period of time, they are broken down together with the mineral contents, which are called clay minerals. These are the fundamental blocks of the type of soil called clay. The kind of rocks from which clay is derived is igneous rock which contains great amount of silicate minerals.

There are three common silicate mineral groups from which clay minerals are derived, to wit:

I. Four aluminosilicate group. This is subdivided into 4 sub-groups.

a) *Pyroxene* - This is a group of complex aluminosilicate mineral, rich in Ca, Mg and Fe. It is dark in color and high in density. The most common member is augite  $[(Ca, Na) MgFeAl] (SiAl_2)O_3$ . Its specific gravity is 3.2 to 3.3 and its hardness is between 5 and 6 on the Moh's scale. It is dark green to black in color and shows a vitreous luster on its cleavage surfaces.

b) *Amphibole*. - This group is similar to pyroxene being a complex silicate containing varying amounts of Ca, Mg, Fe and Al. However, the amphibole formula includes the OH radical which is not present in pyroxene. The most common member is hornblende  $[(CaNa)_2 Mg, Fe, Al, Si, (SiAl, O)_2 (OH)_2]$ .

This is dark-green to black in color with a fibrous texture on its cleavage surfaces.

c) *Mica* - This group includes several silicates of Al in which K is always present, along with the OH ion and in which Mg or Fe may or may not be present. Its hardness is low, on the range from 2.5 to 3 on the Moh's scale. Muscovite is a light-colored mica, commonly known as isinglass  $[(KAl)_2 (AlSi_3O_{10}) (OH)_2]$ . It is characterized by its perfect cleavage in a single set of parallel planes.

d) *Feldspar* - This group consists of Aluminum silicate with one or two of the ions, K, Na or Ca. Hence, there are two groups of feldspar. The alkali feldspar contains Na and/or K while those that contain Na and/or Ca are the plagioclase feldspar.

2. Olivine - This silicate mineral group contains Mg and Fe which are both heavy metals. Hence this group has a high specific gravity ranging from 3.3 to 4.4. The chemical composition is  $(MgFe)_2SiO_4$ . On the Moh's scale, it is 6.5 to 7 in hardness. It has a vitreous luster with a characteristic green color.

3. Quartz - This group consists of silicon dioxide ( $SiO_2$ ). It is a hard mineral (7 on the Moh's scale). It breaks with a conchoidal fracture, so that broken pieces of crystalline quartz resemble massive chunks of broken glass. Colorless quartz is called rock crystal. Amethyst is violet variety. Rose quartz is rose-red or pink. The specific gravity of quartz is 2.65.



The outer points of the octahedron, may also be occupied by OH ions (hydroxide). The octahedrons are arranged in sheets in which the oxygen or hydroxide ions are shared to form a strongly bonded layer.

The clay minerals are characterized by alternation of tetrahedral sheets with octahedral sheets, bounded by sharing of the oxygen ions making up the free points of the silicon-oxygen tetrahedrons. Two or three such sheets bounded together form the unit layer of the clay mineral.

Two kinds of sheet combinations are illustrated as follows:

One kind of sheet combination is a unit layer formed by one tetrahedral sheet bonded to one octohedral sheet. This arrangement is designated by the symbol 1:1 (Figure 3).

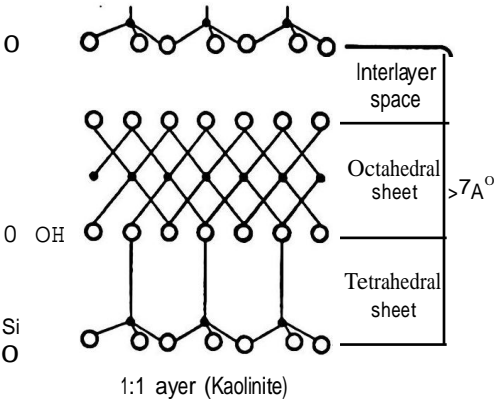


FIGURE 3 SCHEMATIC SIDE VIEW OF LATTICE LAYER STRUCTURE SHOWING 1:1 COMBINATION

The second kind of sheet arrangement consists of one octohedral sheet between two tetrahedral sheets. This arrangement is designated by the symbol 2:1 (Figure 4).

Stached unit layers, **one** above the next, form the total mineral structure. Between the lattice layer groups is the interlayer space, which maybe unoccupied or maybe occupied by layers of bonded base cations, by hydroxide ions or by water molecules.

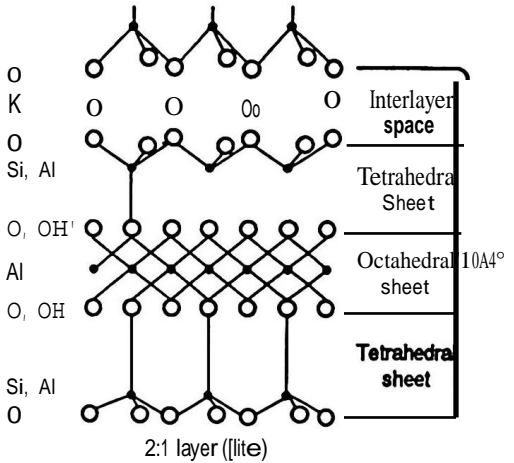


FIGURE 4. SCHEMATIC SIDE VIEW OF LATTICE LAYER STRUCTURE SHOWING 2:1 COMBINATION

### CLAY MINERAL GROUPS

Clay minerals are referred to by group names. The crystalline groups are montmorillonite, illite, vermiculite, chlorite and kaolinite groups and are referred to as silicate clays. The non-crystalline or amorphous clays also contain silica Sesquioxides **are a** group of various iron oxides and titanium oxide clays.

Montmorillonites are the swelling and sticky clays. They are referred to as 2:1 type or expanding lattice (Figure 4). In this type of clay, water easily penetrates between layers causing the clay particles to swell. It is of the expanding type. If the solution contains mostly Na, the clay may swell 3 to 10 times its dry volume and become like gels. Soils of

the arid regions and poorly drained soils developed from alkaline parent rocks such as limestone mostly have montmorillonite clays. Bentonite an impure deposit of montmorillonite, is used to seal earthen ponds, as solution stiffeners or as thickeners in paints and lipsticks. The general formula of montmorillonite clay..mineral is  $\text{OH} \rightarrow \text{Al, Si, O, NH, O,}$

Illites have gross structures similar to that of the montmorillonites, that is a 2:1 type of clay of silica and alumina sheets. However, this type of clay has larger  $\text{K}^+$  holding adjacent layers held together so tightly that water can not penetrate between layers. Thus, illites have slight to moderate swelling depending upon how many of the planes of  $\text{K}^+$  have been weathered out allowing some clay layers to be separated and the clay to expand somewhat like montmorillonite. Illite, a potassium aluminum silicate compound has the formula  $\text{KAl}_2\text{Si}_2\text{O}_{10}(\text{OH})$ .

Vermiculites are similar in structure to illites but the layers are held more weakly together by hydrated  $\text{Mg}^{2+}$  rather than tightly together by  $\text{K}^+$ . Thus vermiculites have some swelling but not as much as montmorillonites. They exhibit a high cation exchange capacity.

Chlorites are common in some soils. They are often called 2:2 type clays because they are similar to the unit lattice of vermiculite, except that the hydrated Mg in vermiculites is a firmly bonded  $\text{Mg}(\text{OH})_2$  octahedral sheet. A layer of chlorite clays has 2 silica tetrahedra, an alumina tetrahedra and a magnesium octahedra sheet (2:2). Chlorites do not swell when wetted (non-expanding) and have low cation capacities.

**Kaolinites** are commonly distributed clay minerals. A kaolinite has a sheet of silica tetrahedron per sheet of alumina tetrahedron per layer (1:1 type). It is illustrated in Figure 3.

Almost no substitution of  $\text{Al}^{3+}$  for  $\text{Si}^{4+}$  or  $\text{Mg}^{2+}$  for  $\text{Al}^{3+}$  has occurred in kaolinite. Hence, the net negative charge or cation exchange capacity is low. However, each layer

has one plane of  $\text{O}$  replaced by  $\text{OH}^-$ . This results in strong Hydrogen ( $-\text{H}-$ ) bonds to the oxygen plane of the adjacent layer. Kaolinites have strong hydrogen bonding capacity such that they do not allow water to penetrate between the layers and have almost no swelling (non-expanding type). The chemical formula of kaolinite molecule is  $\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$ . These are the types of clays used for pottery work because they do not shrink and swell.

Amorphous clays are mixtures of silica and alumina that have not formed well-oriented crystals. Mixtures of other weathered oxides (iron oxide) maybe a part of the mixture. Typically, these clays occur where a large amounts of weathered products existed but did not have the condition or time for good crystal formation. Amorphous clays are not well characterized but exist in many soils in varying amounts. Their properties are unusual having high positive charges and hence have high cation exchange capacities. These clays have a variable charge that depend on how much  $\text{H}^+$  is in solution.

Sesquioxides are mixtures of  $\text{Al}(\text{OH})_3$  and  $\text{Fe}_2\text{O}_3$ . These types of clay are formed as a result of continuous leaching by rainfall and long-time intensive weathering of minerals in humid, warm climates when most of the silica and alumina are washed away. The remnant materials are the sesquioxides. They have low solubilities. Sesquioxide clays refer to the clay soils of Fe and Al because of their formulas written as  $\text{Al}_2\text{O}_3 \cdot x\text{H}_2\text{O}$  and  $\text{Fe}_2\text{O}_3 \cdot x\text{H}_2\text{O}$ . These clays can be either amorphous or crystalline. Small amounts of these clays exist in many soils, particularly in the humid, well-drained soils of tropical areas where intense weathering has occurred for thousands of years. Iron oxide and iron hydrate commonly color the soils various shades of red to yellow. These clays do not swell, are not sticky and do not behave as do the silicate clays.

**Mixed Layer Combinations.** Many clay materials are composed of more than one clay minerals. These clay mineral components are

random mixtures of discrete units, so that there is no preferred geometric orientation. Mixed layers of illite and montmorillonite and of chlorite and vernalite are common in clay minerals and can only be detected by careful x-ray diffraction techniques.

## DIFFERENT TYPES OF CLAY

According to the manner of formation, clay is classified as residual clay and **sedimentary clay**. Residual clay is derived directly from the weathering of rocks into fine particles which are mixed with water and materials from the surrounding soil. Sedimentary clay is formed when particles of weathered rocks are carried from the place where they were formed, usually by streams of water and deposited in another place. It occurs in layers or sheets (Figures 3 and 4).

According to their chemical composition and properties, clay may be classified as follows:

**Kaolin or China Clay** - is white in color, consisting chiefly of the mineral, kaolinite. It is **very fine** in particle size with a **hardness** of 2.0-2.5 on the MOHs scale. It is chemically inert.

It is used in the paper industry, for making paper products smoother, whiter and more printable. In ceramics, it is used in making **whitewares**, special refractories and porcelain insulators. In the manufacture of natural and synthetic rubber, kaolin is used as a **filler**, adding strength, abrasion resistance and rigidity to the product. It is also used in the making of plastics and insecticides and in the refining of petroleum.

Calcined Clay, a commercial product of kaolin is used in the making of paint, ceramics, plastics, paper and other materials.

Ball Clay is darker in color than kaolin or china clay. It is composed mainly of kaolinite with another clay mineral, montmorillonite and small amount of organic

matter. It has finer grain-particles than China clay. It is also more plastic and stronger than China clay. It is used in the production of white wares and sanitary wares.

**Fire clay.** This type of clay can withstand a temperature of 1500C or higher. It is primarily composed of kaolinite with minor amounts of illite and quartz. It is light to dark gray in color.

The foundry industry uses fire clay to bind sands into shapes in which metals can be cast.

**Fiat Clay.** This is a special type of fine clay which is non-plastic and is very hard. It is used in the refractory industry.

**Diaspore Clay.** This is also a special type of clay that is composed of the minerals, diaspore and kaolinite. Diaspore is hydrated aluminum oxide (85%  $Al_2O_3$  and 15%  $H_2O$ ). This mineral is quite hard (6.5-7 on MOHs scale) and very refractive. Hence it is used in making refractory brick.

**Bentonite.** This clay is mainly composed of the clay mineral, montmorillonite and is formed by the alteration of volcanic ash. This is used as drilling muds and as catalyst in the petroleum industry and as **bonding** clay in foundries. It is also used as bleaching clay to remove coloring matter from oil and as absorbent for oils, insecticides, alkaloids, vitamins and many other materials.

**Attapulgite Clay.** This clay is made up of hydrated magnesium **aluminum silicate** and is commonly known as **fullers earth**. It is used for the neutralization, deodorization and refining of mineral and vegetable oils, fats and waxes as well as in drilling muds for salt-water formation. It is also used as catalyst and adsorbent for alkaloids, vitamins, carbohydrates and many other compounds.

**Miscellaneous Clay.** This is a mixture of different proportions of illite, chlorite,

kaolinite and montmorillonite plus a variety of non clay materials. It is largely used in the clay product industry i.e., in the manufacture of bricks, tiles, sewer pipes, terra cotta and other items. The properties of this clay are dependent on its composition and the particle size of the constituents. Sandy clay and clayloam belong to this category.

According to their location, clays maybe classified as surface clays, alluvial clays and bedded clays.

**Surface Clays.** The most abundant clay is that appearing at or very near the earth's surface. In its natural state it may be red, brown, yellow, green, blue or black. Upon firing, these clays are red or red brown. Typically, surface clays are too plastic to use in making clay products without adding sand. They harden at temperatures from 1350 to 1650°F. These clays are used by the first potters and are still used in making cooking pots, flower pots, stoves, ovens, tiles and art wares.

**Alluvial Clays.** These clays are sedimentary clays deposited at the surface of rivers or in lakes or in ponds. They are very plastic. Cooking wares, art wares, bricks and tiles can be made from these clays.

**Bedded Clays.** These clays are found along rivers where the water is worn deeply through the banks. From the top to the bottom of the bank are layers of surface clays, sand and gravel and near the bottom are the bedded clays. They appear in stratified formation due to their layers of depositions. Under heat and pressure, the layers have become firmly pressed together. They are very plastic due to their fine grains. They vitrify at approximately 2000°F and "become very strong. Good quality jars, bricks and floor tiles can be made from these clays. Bedded clays can be seen when digging wells or other deep excavations.

## GENERAL PROPERTIES OF CLAY MINERALS

The varied properties of the clay minerals govern their economic uses. Some of these properties are as follows:

### Ion Exchange

This refers to the ability of clay minerals to hold certain cations and anions which are readily exchangeable for other cations and anions. The most common exchangeable cations in clay are Ca, Mg<sup>++</sup>, H<sup>+</sup>, K, NH<sub>4</sub><sup>+</sup> and Na<sup>+</sup>. The more common exchangeable anions are SO<sub>4</sub><sup>-</sup>, Cl, PO<sub>4</sub><sup>-</sup> and NO<sub>3</sub><sup>-</sup>.

There are two major causes for the exchange capacity of the clay minerals. Broken bonds around the edges of the silica-aluminum units give rise to unsatisfied charges which are balanced by absorbed cations. Substitution within the structure results in unbalanced charges satisfied by exchangeable cations. In some cases, some anions maybe absorbed by replacement with exposed OH. This property of ion exchange is important in fields where clay materials are used. In soils, the retention and availability of potash added in fertilizers depend on cation exchange between the potash salt and the clay mineral in the soil.

Among the clay minerals, montmorillonite has a high cation exchange capacity (CEC), illite, an intermediate CEC and kaolinite, a low CEC. The hydrous sesquioxides of iron and aluminum have a very low CEC.

Montmorillonite-->Illite-->Kaolinite-->Sesquioxides of Al & Fe

Soils that are developed from recently formed regolith tends to be rich in montmorillonite and illite. As time passes and the clay minerals undergo further changes, kaolinite becomes dominant/ In old soils, there remains little except the sesquioxides of Al and Fe. Soils of low cation - exchange capacity with low proportion of base cations are poor agricultural soils while soils of high cation exchanges capacity are naturally rich agricultural soils.

## Clay Water System

Another important property of clay minerals is their ability to hold water. This is the water in pores, on the surfaces and around the edges of the minerals composing the material. It is also the interlayer water between the unit cell layers of the minerals, such as montmorillonite. The presence and volume of water held by clay minerals determine the plasticity, bonding capacity, suspension, compaction and other properties of clay, which in turn, govern their economic uses.

## Dehydration

When clay minerals are heated, dehydration occurs. Thus, there is a loss of water held by the clay minerals. The heating of clay materials also causes changes in the clay-mineral structures. At relatively high temperature these structural changes facilitate the formation of new mineral phases. These structural modifications are particularly important in the firing of clay materials,

## Clay-Mineral Organic Reactions

Another property of clay minerals is their ability to react with organic materials. Clays with a high absorbing capacity are used in decolorizing oils while others provide catalysts in the cracking of organic compounds.

## PROPERTIES OF CLAY

The properties of clay are largely dependent on the chemical composition of the clay, that is the type of clay minerals composing it and the particle size of the constituents. The common properties are as follows:

**Plasticity.** Clay is the only material that possesses plasticity in its natural state. Most clays become plastic when mixed with varying proportions of water, i.e. clays have the ability

to undergo permanent deformation in any direction without being ruptured under a stress. Clays range from those which are very plastic called fat clay to those which are barely plastic called lean clay. The type of clay minerals; the particle size **and shape and** the amount and type of non clay minerals **are the** factors that affect the plastic property of clay.

**Strength.** Clays are characterized by having such strength, such that structural clay products can maintain their desired **shape**. Green strength is the strength of a clay material in the wet, plastic state while dry strength is the strength of the clay after it has been dried.

The strength of clay also depends upon the same variables that affect plasticity. Dry strength is dependent on the proportions of fine particles present in the clay, the shape of the individual particles, the degree of hydration of the colloidal fraction and the extent of drying. The presence of a small amount of montmorillonite, which is of very fine size and highly hydrated generally increases the dry strength of the clay material.

**Shrinkage.** This refers to the loss in the volume of clay when it is dried or when it is fired, when used for structural clay products. This is dependent on the water content; the character of the clay materials and the particles size of the constituents. Drying shrinkage is high in very plastic clays and tends to produce cracking and warping. It is low in sandy clays or clays of low plasticity and tends to produce a weak, porous body. The presence of 10-25% montmorillonite in the clay material causes excessive shrinkage, cracking and slow drying.

**Color.** Some clays are almost pure white, because they consist wholly of white clay or kaolinite. Other clays are of various shades of yellow, brown or red due to the presence of iron minerals while other clays are nearly black due to their organic content.



**Size.** Based on Wentworth scale, claystone consists largely of particles of clay grade smaller than 0.004 mm (4 microns). Clay materials have the smallest particle size compared to sand and silt.

**Vitrification.** The temperature range of glass formation or vitrification is a very important property in the production of clay products. Vitrification is due to a process of gradual fusion in which some of the more easily melted constituents begin to produce an increasing amount of liquid which makes up the glossy bonding material in the fired product. Kaolinites have higher vitrification temperature than illites, chlorites and montmorillonites.

## VIGAN CLAY SAMPLES

Some properties of Vigan clay soil samples collected in **Barangays** Mindoro and Salindeg are presented in Tables I and 2. The pH values of all the samples are acidic, with a pH of less than 7. Bulala soil samples are noted to be more acidic than Mindoro soil samples. The two Mindoro soil

samples have pH values of 6.3 and 6.5 respectively while the four Bulala soil samples have 5.5, 5.4, 5.6 and 5.5 pH values respectively. These data indicate that more **H** and **Al** are found in the latter than in the Mindoro soil samples, since these two ions are the principal acid-generating cations in most soil.

Among the four basic cations ( $\text{Ca}^{++}$ ,  $\text{Mg}^{++}$ ,  $\text{Na}^+$  and  $\text{K}^+$ ),  $\text{Ca}^{++}$  predominate in all the soil samples. Samples from Bulala contain more of these ions than Mindoro samples. This is an indication that Bulala soil samples have greater capability to hold and exchange cations than the soil samples than Mindoro samples. This is confirmed by the Cation Exchange Capacity Value presented in Table 1. The CEC values of the four Bulala soil samples are 27.2, 25.7, 23.8 and 33.4 respectively while CEC values of Mindoro soil samples are 8.0 and 9.7 respectively. The soil samples from Bulala have a higher degree of chemical activity than the Mindoro soil samples. This shows that Bulala soil samples contain more of such clay minerals as montmorillonite and illite, than kaolinite while Mindoro soil samples consist more of kaolinite and hydrous sesquioxides of iron and aluminum.

**Table 1. Chemical Analysis of Clay Soil Samples From Barangays Mindoro and Bulala, Vigan, Iloos Sur**

Soil Samples	PH	Milliequivalents/100 gms/soil Exchangeable Bases				Summ.	Exchange-able Acid	CEC Summary
		Ca	Mg	Na	K			
<b>Mindoro Samples</b>								
1	6.3	2.8	0.4			3.7	4.8	8.0
2	6.5	2.3	1.4			3.8	5.9	9.7
<b>Bulala Samples</b>								
1	5.5	9.2	2.2	0.2	0.1	11.7	15.5	27.2
2	5.4	10.0	4.8	0.7	0.1	15.6	10.1	25.7
3	5.6	7.8	7.6	0.2	0.1	15.7	8.1	23.8
4	5.5	12.4	9.3	0.4	0.1	22.2	11.2	33.4

• Taken from the Records of the Department of Agriculture Office, Vigan, Ilocos Sur  
Cation exchange capacity

**Table 2. Physical Analyses of Texture of Clay Soil Samples from Barangaya Mindoro and Bulala, Vigan, Ilocos Sur**

Soil Samples	Sand	Silt	Clay
<b>Mindoro Samples</b>			
1	<b>90.2</b>	1.2	<b>8.6</b>
2	<b>88.2</b>	2.2	<b>9.6</b>
<b>Bulala samples</b>			
1	38.2	30.0	<b>31.8</b>
2	33.2	30.0	<b>31.8</b>
3	33.4	26.0	40.6
4	22.2	<b>24.0</b>	33.8

Taken from the Records of the Department of Agriculture Office, Vigan, Ilocos Sur

Table 2 shows the percentage composition of clay, as well as sand and silt in the soil samples. Mindoro soil samples consist mostly of sand, (90.2% and 88.2% respectively), while only 8.6% and 9.6% of the samples are made up of clay.

The predominant component of Bulala soil samples is clay. The four samples contain 31.8%, 39.8%, 40.6% and 33.8% clay materials respectively. The only sample that contains more sand is sample one which contains 6.4% more of sand than clay.

The clay deposits therefore in Barangays Mindoro and Bulala belong to the variety of miscellaneous clay. These deposits are made up of mixtures of different kinds of clay minerals with the addition of non-clay materials. This variety of clay is used as raw materials in the manufacture of clay product like bricks, pots, tiles and pipes.

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