DE NATURA LUTORUM*

by

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ABSTRACT

Despite recent advances in clay mineralogy there is still no generally accepted definition of the terms *clays* and *clay minerals*. This problem is discussed from the historical aspect: the similarities of definitions given from the 16th century to the present are remarkable and lead one to consider whether possible simple modifications might not eventuate in some measure of universal agreement. Comparison of nomenclature and classification schemes shows that from the time of Theophrastus to that of Agricola there appears to have been little advance, but a change becomes noticeable by the end of the 18th century, and by the end of the 19th old names, such as porcelain clay, etc., have given place to what are now regarded as names of mineral species or varieties. Only after the 1920's have satisfactory groupings been obtained, but no internationally acceptable classification has yet been devised. Finally, the various factors operative during clay formation by weathering and diagenesis are discussed, and it is stressed that field and laboratory studies are complementary.

INTRODUCTION

From early times there has been a desire by those interested in *earths* and *clays* to devise some definitions and some system of classification, but until the advent of X-ray diffraction studies in the twenties the difficulties encountered were almost insuperable. This is clearly evident from earlier books on mineralogy where the arrangements are many and varied and very few agree in classifying the same clays under similar headings. The advent of modern methods, however, has led to even greater difficulties in that names have been indiscriminately given to minerals which are only trivial varieties and consequently during the last decade or so there has been a considerable upsurge of interest in the problem of nomenclature and classification and there has been a clearly manifest desire to devise a system which would be internationally acceptable.

Those of us who have been particularly interested in and aware of this problem have inevitably had to consider more deeply the origins of our science and the fundamental properties of the materials with which we work. In

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*From *lutum*—clay. *Argilla*, which indicates specifically a white clay, and *creta*, which can also signify chalk, are unsuitable here. The most usual term used by the ancients was *terra*, but this has rather a wider significance than that intended. *Lutum* appears to have been used for clay as distinct from "earth" – see Pliny, *Nat. Hist.*, XXXV, 169.

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consequence, I should like here to consider very briefly some of the older ideas and how these have developed into our present concepts.

The phrase "Clays and Clay Minerals" is an integral part of the title of this annual conference and it seems fitting to consider these two basic concepts first before proceeding to deal generally with their origin.

WHAT IS CLAY?

Clays are familiar to most scientists—and laymen—but the significance attached to the word depends upon the discipline involved. Thus the potter, the farmer, the engineer, the geologist and the mineralogist would not define clay in the same manner although there is fundamentally much in common among the materials visualized.

Among the ancients, clays were included under the rather wider term of *earths*, and earth was regarded as one of the four elements, having dry–cold properties. It was distinguished from the stones by its friable nature. But probably the earliest actual definition is that of Agricola (1546) who stated that an earth is:

"a simple mineral body which can be worked in the hands when it is moistened and from which mud can be made when it is saturated with water"

—a definition that still persists as evidenced by that given by Grim (1953):

"In general the term clay implies a natural, earthy, fine-grained material which develops plasticity when mixed with a limited amount of water",

which is virtually that of Agricola rendered into modern idiom. Similar definitions may be traced through the intervening period, e.g. Geikie (1886) comments that:

"Clay [is] a fine grained argillaceous substance . . . [which] usually contains interstitial water and when wetted can be kneaded between the fingers; when dry it is soft and friable and adheres to the tongue. Shaken with water it becomes mud, and even a small quantity will make a glass of water turbid, so fine are the particles of which it is composed".

Dana (1888) defines clay as:

"a soft, impalpable, more or less plastic material",

and Geikie (1905) remarks that:

"clays are aggregates of very finely divided mineral matter which become plastic when moistened".

In most of these definitions the fineness of the particles is stressed. One of the earliest to associate a definite particle-size range with clays was Kirwan (1794), who comments that:

"[Clays] are for the most part diffusible in water, and do not immediately sink in it as sands do: or if so compact as immediately to sink, they soften, crumble or moulder away in it, some sooner, some later, either to a ductile viscid pulp, or to a powder. . . . They harden when heated, scarce ever effervesce with acids and are difficultly soluble in them. . . . The constituent parts of clays are argill [alumina] and siliceous stones from the size of 1/100 [*ca.* 0.25 mm] to 1/1000000 [*ca.* 0.025μ] of an

inch. All other ingredients (except water), and there are many, are extraneous to its composition."

One may compare with this the remarks of Merrill (1906) that:

"About the only feature characteristic of all clays is that of plasticity, when wet, and this is dependent, apparently, upon . . . the size and shape of the individual particles, and in some cases at least the presence of colloidal matter",

and of Chukhrov (1955) that:

"So-called clays are plastic aluminosilicate rocks, in which the predominant mass of particles has a diameter less than 0.01 mm. . . . Mineral particles in clays (as in soils) have different sizes, i.e. clays are polydisperse rocks."

The property of clays of hardening on heating is noted more particularly in definitions by ceramists, e.g. the American Ceramic Society definition (Searle and Grimshaw, 1959) is:

"Clay is a fine-grained rock which, when suitably crushed and pulverised, becomes plastic when wet, leather-hard when dried and on firing is converted to a permanent rock-like mass",

but it has also been remarked upon by mineralogists and geologists, as is clear from the quotation above from Kirwan (1794) and the definition given by Holmes (1928) in which he observes that clays

"become hard and stone-like on being heated to redness."

Further complications arise from the use of *clay* as a specific particle-size term, although when it is used in this sense it is preferable to be specific and to state the *clay fraction*—i.e. that fraction of the material the particles of which are below a specific equivalent spherical diameter (e.s.d.). The size generally agreed upon as the upper limit of e.s.d. for the clay fraction of soils is 2μ, but there are still many inconsistencies in sedimentary petrology. Pettijohn (1957) and Müller (1961) compare and contrast several scales which have been suggested, but at present the Wentworth (1922) scale, using 1/256 mm (3.9μ), is probably the most generally used. Recent publications, however, use values from 20μ to 2μ, e.g. Correns (1939) and Füchtbauer (1959) suggest 20μ, Seidov (1959) and Konta (1960) argue on the basis of 10μ, Rukhin *et al.* (1958) use 5μ, Pettijohn (1957) recommends 3.9μ or 2μ, Shepard (1954) uses 3.9μ, and Sedletskii (1945), Lemcke, von Engelhardt and Füchtbauer (1953), and Müller (1961) employ 2μ. In addition, Chukhrov (1955) quotes Melnikov (1949) as noting that many properties of clay-forming particles (hygroscopicity, plasticity, etc.) show an abrupt change at a diameter of 1μ—which is the approximate usual upper limit of size for particles considered as colloids (Kruyt, 1952).

If one then considers the definition of clay in terms of the amount of clay fraction in the rock or soil, confusion becomes worse confounded as is evident from the three triangular diagrams shown in Fig. 1, on which the areas covering *clays* are shaded—nor do these areas bear any specific relationship to the e.s.d. taken as the upper limit of size of the clay fraction. It will be evident, therefore, that a considerable amount of confusion in sedimentary petrology has still to be internationally obviated. In pedology, on the other

hand, it is generally internationally agreed that the diagram shown in Fig. 1(c) is acceptable. Pettijohn (1957) remarks that:

"The definition based on grain size is least satisfactory because most commercial clays are not clays by this definition: ... [Furthermore] the term clay as applied to aggregates may contain as little as 50% of clay sized material or it may be defined as containing no less than 80%."
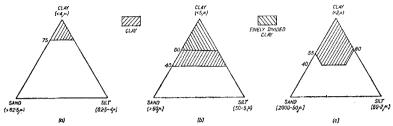

FIGURE 1.—Triangular diagrams showing compositions designated as *clay* by: (a) Pustovalov (see Rukhin *et al.*, 1958) and Shepard (1954); (b) Rukhin *et al.* (1958); (c) Soil Survey Staff (1951).

Many definitions of clays from the time of Kirwan (1794) refer to the chemical consitution as consisting largely of silica, alumina and water (e.g. Tschermak, 1888) or of hydrated aluminous silicates (e.g. Geikie, 1886; Dana, 1888; Merrill, 1906; Holmes, 1928; Pettijohn, 1957). Such chemical criteria, however, are not particularly convincing, since well-defined clay minerals such as nontronite, saponite, and glauconite are low in alumina, and bauxite, which is regarded as a clay commercially, has virtually no silica.

From the above discussion it will be clear that chemical composition can be largely discounted and that the majority of definitions of clay so far given are based on one or more of the following physical criteria, namely:

(a) plasticity;
(b) small particle size;
(c) hardening on firing.

The first of these is open to objection in that, as Grim (1953) remarks, it would exclude such materials as flint-clays—and, furthermore, plasticity is a property of most, if not all, very fine materials, e.g. quartz (Geikie, 1905). Particle size can be objected to on the grounds that clays are, as Chukhrov (1955) remarks, polydisperse rocks or soils, that many commercial clays would be excluded (Pettijohn, 1957), and that there is no unanimity as to where the upper limit of size for the clay fraction ought to be drawn, nor to the minimum amount of clay fraction necessary to constitute a clay. Hardening on firing might be objected to on the basis of the presence or absence of fluxes and all clays would not become hard on simple heating to redness.

Yet if a material is a natural earthy deposit satisfying two out of three of these criteria it would, in most disciplines, be classed as a clay.*

* Possible exceptions would seem to be certain marls which can be of very fine particle size and plastic, but here a simple chemical test would differentiate.

WHAT IS A CLAY MINERAL?

Just as clays are difficult to define, so are clay minerals. In early books on mineralogy the recognized crystalline ones, such as kaolinite, were usually considered as separate species and the clay family, or clay group, was classed separately (e.g. Walchner, 1829). Tschermak (1888) remarks that:

> "[The clay group consists of] soft, for the most part earthy, crystalline and amorphous minerals, which usually contain only silica, alumina and water."

In addition it was often considered that clays were mixtures of one essential mineral with varying amounts of impurity, e.g. as late as 1929, Blanck stated that:

> "Clays are mixtures of kaolin with microscopical amorphous-appearing masses of similar composition and with SiO_2- and Al_2O_3-hydrogels",

although even in 1906 Merrill regarded this as an "unfounded assumption" and Milner (1929) pointed out that kaolinite does not play the "supreme part" in clays: indeed Ross and co-workers (see Ross, 1927) had earlier shown that many clays did not in fact contain kaolinite. The supposed essential constituent was also regarded as "Thonsubstanz" (Senft, 1867), "la véritable argile" (Brongniart and Malaguti,* 1841) and "clayite" (Mellor, 1908); the last two terms are, however, virtually synonymous with kaolinite since the authors were concerned chiefly with china clays and only extended their observations to other clays by inference. These concepts have been reviewed by Grim (1953) and it would be superfluous to go into further detail here.

It is certainly not adequate to define a clay mineral as "any mineral which occurs in a clay" since not only does the indeterminate nature of the word *clay* militate against such a definition, but it would include many accessory minerals which are not characteristic. Furthermore, we must appreciate that clay, or rather the clay fraction of a soil or rock, frequently consists of a mixture of crystalline and amorphous material. This idea has been mentioned in many of the early definitions of clays, but with the advent of refined techniques, it has become customary to consider the clay fraction as consisting almost entirely, if not entirely, of crystalline compounds—a concept which is indeed stressed by Grim (1953). More recent work, however, has led to the conclusion that the pendulum swung too far in this direction and the researches of, *inter alia*, Sudo (1954), Fieldes (1957), and Mitchell and Farmer (1962), which show the wide distribution in clays of material amorphous to X-rays, lead us back to the older concept.

In 1958 an attempt was made to produce a definition of a *crystalline* clay mineral which might be generally acceptable and this was later published for general criticism. It reads (Mackenzie, 1959):

> "Crystalline clay minerals are hydrated silicates with layer or chain lattices consisting of sheets of silica tetrahedra arranged in hexagonal form, condensed with octahedral sheets; they are usually of small particle size."

* It is interesting to note that these authors boiled their clays for $1-1\frac{1}{2}$ min with a solution of KOH to determine the amount of contaminating "gelatinous silica" (cf. Foster. 1953: Glenn *et al.*, 1960).

This left unstated the definition of amorphous clay minerals, but arguing from the classification given by Brown (1955) these could well be defined as the

"materials in the clay fraction not shown by the method of investigation to be crystalline".

The definition of crystalline clay minerals was reconsidered, in the light of criticism, at an international meeting in Copenhagen in 1960 when it was suggested that there be added to the above:

". . . and they have the capacity to form in combination with water more or less plastic aggregates".

Such an addition, to the author, does not seem to be helpful since, as mentioned above, probably all fine-grained materials show plasticity to some extent.

This type of definition has been objected to in some quarters as being essentially that of phyllosilicates in general rather than of clay minerals, and although the majority of clay minerals are phyllosilicates, the converse, that all phyllosilicates are clay minerals, is by no means true. In fact, it is undoubtedly correct, as mentioned by Grigorev (1961), that clay minerals belong to one or another of the ordinary mineral species. Consequently, one wonders whether, rather than define a clay mineral strictly, it might not be better to give a very general definition such as

The minerals which normally predominate in the colloidal fraction of clay rocks, soils, etc.
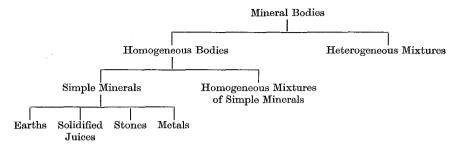
Such a definition does not separate clay minerals from classical mineralogy, but does appear to give adequate coverage, and, by insertion of the word "normally", to exclude accessories.

CLASSIFICATION OF CLAY MINERALS

Earths were usually classified by the ancients on the basis of their uses and named after the locality from which they originated, e.g. Kimolian, Melian, Lemnian, etc. (Theophrastus, ca. 300 B.C.), or from some specific characteristic, e.g. argilla—a white clay, saxum—a hard clay, probably montmorillonite (Robertson, 1949), sigillim caprae—another name for Lemnian earth, arising from the figure of a goat with which the lumps were stamped (Agricola, 1546), etc. This system, while undoubtedly useful in the industries then extant, did not lend itself to strict characterization but remained the only basis of classification until relatively recent times. And, of course, factual mineralogy was well interlarded with fables.

The first serious classification of minerals is due to Georgius Agricola (1546) who, in De Natura Fossilium, brought the subject "down to earth", so to speak, and devised the scheme:*

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* Based on the notes of Hoover and Hoover in De Re Metallica (Agricola, 1556) and Bandy and Bandy in De Natura Fossilium (Agricola, 1546).

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                        Mineral Bodies
                             |
        ┌────────────────────┴────────────────────┐
  Homogeneous Bodies                      Heterogeneous Mixtures
        |
   ┌────┴──────────────┐
Simple Minerals    Homogeneous Mixtures
   |                of Simple Minerals
┌──┬──────┬─────┬──────┐
Earths Solidified Stones Metals
       Juices
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In this volume one "book" out of the ten it contains is devoted to *earths*, the majority of which are clays, but the nomenclature had changed little from the time of Theophrastus.

Unfortunately, such nomenclature is of relatively little use now since it is doubtful whether most of the *earths* were monomineralic and one name could be used in two contexts—e.g. Melian earth could be either a siliceous kaolin or a fairly pure, very fine silica, since deposits of both these types are found in Melos (Robertson, 1958). The fact that this system of nomenclature and classification led to some anomalies was appreciated by Agricola (1546) who states:

> "I accept the common practice of giving locality names to earths . . . [but] I believe that earths from one locality may be worthless, while similar ones from another are valuable",

and over 200 years later by Kirwan (1794):

> "As earths originally received their denominations from external and not from internal characters, they cannot now, without creating much confusion, be arranged according to their real specific properties."

Despite the limitations imposed by such primitive nomenclature and classification it is certain that the ancients could distinguish, for example, predominantly kaolinitic from predominantly montmorillonitic clays (Theophrastus, *ca.* 300 B.C.; Roberston, 1958). Similarly, Kirwan (1794) devotes four pages to the distinction of the various types covered in his "Argillaceous Genus"[*] from each other. Although differentiation was based on such superficial features as fracture, friability, hardness, unctuousness, adhesion to the tongue, opacity, etc., it is clear that a considerable measure of distinction was available at this time, but it had probably advanced only in detail over that appreciated by the ancients.

Walchner (1829), like Kirwan, employed a chemical classification, and his nomenclature is essentially similar to that of Kirwan (1794), although incorporating minerals discovered in the interim—e.g. allophane (Stromeyer, 1816), which is included under the "aluminum" family. Dufrénoy (1856) also

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[*] This included only aluminous minerals, and magnesian minerals, such as "meerschaum" and "serpentine", were considered separately under "Muriatic Genus".

employed a chemical classification and distinguished clays formed *in situ* from those formed by "dépôt chimique" but Breithaupt (1836–47) in the interval laid considerable stress on physical properties in his classification and lists clays under "Porodini (Guhren)". At about this period many of the minerals we now recognize were named—e.g. halloysite and nontronite by Berthier (1826, 1827) and montmorillonite by Mauduyt (1847)—and consequently each successive "mineralogy" has an accretion of new names.

The fact that there was still considerable confusion is, however, evidenced by the history of montmorillonite. The original material from Montmorillon, France, was named "montmorillonniste" by Mauduyt (1847), who simply stated that it contained silica, alumina and water. This material was analysed in the same year by Damour and Salvétat (1847), who, on comparison with the analysis of a reputed "halloysite" from Confolens described by Berthier, renamed the mineral "halloysite". The name "montmorillonite", or rather the German form "Montmorillonit", appears to have been used first by Naumann in 1850,* but despite this Salvétat in 1851 described as "halloysites" some other minerals which from their properties and analyses† appear to be essentially montmorillonites; he also names a mineral from Condé, which from its analysis appears to be halloysite, "smectite". This is the only instance the author has been able to find of the application of the name smectite to a mineral not of the montmorillonite group, for which, as fuller's earths, the term "smectis" has been used since early times. This error would undoubtedly have been avoided had Damour and Salvétat (1847) appreciated that their type material from Confolens—later called confolensite by Dufrénoy (1856)—was not identical with the original halloysite of Berthier (1826) from Angleur, but that Mauduyt's (1847) identification of the new mineral "montmorillonniste" was correct and that their attribution of names ought to be reversed. The fact that montmorillonite and confolensite were distinct from halloysite was not, however, appreciated for many years to come.

Nevertheless, Damour and Salvétat (1847) comment:

> "It is very probable, as many mineralogists have observed, that [in] minerals, near to clays, . . . the silica, alumina, and water, which are essential constituents, can, in effect, unite in various proportions; it would then be interesting to study . . . the composition of aluminous hydrosilicates . . . which, in general, are met with quite frequently in the mineral kingdom."

In 1887 Le Chatelier attempted to classify clay minerals on the basis of their dehydroxylation temperatures, but this attempt does not seem to have been developed by mineralogists generally.

As an example of the variety in classification systems evident during the 19th century, one may compare those offered in the same year by Dana (1888) and Tschermak (1888). Although Dana records many more trivial

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* The author is indebted to Dr. M. H. Hey for this reference.
† Salvétat (1851) used dilute sodium carbonate to determine the amount of "gelatinous silica" contaminating his clays in place of the dilute sodium hydroxide which he employed in 1847.

names than Tschermak if one omits doubtful species and considers only those
which we know to be distinct species or varieties, the classification of Tscher-
mak is probably the more logical, since Dana, for example, separates glaucon-
ite from the hydro-mica group and halloysite from kaolinite. Nevertheless,
the indications by Dana (1888) of synonymy and variety are, in most instances,
still valid.

Hintze (1897) classified well-crystallized clay minerals under two groups,
the serpentine group and the kaolin group—and in addition gave under
"hydrated aluminium silicates" lists of what he terms "Thon-artige Gemen-
gen" with three groups under the names halloysite, allophane, and mont-
morillonite. He also comments that kaolin and pyrophyllite differ from anti-
gorite and talc by replacement of aluminum by magnesium.

Comparison of the nomenclature and classification of clays by Kirwan
(1794) and of Dana (1894) (Table 1) gives some impression of the advances
which were made during the 19th century. The main difference in the two
classifications is the development of species names, although undoubtedly
many of these species names referred to mixtures or were synonymous of
other species names rather than distinct minerals. The 19th century, therefore,
saw a very considerable development of clay mineralogy in the recognition
of many distinct species and the gradual disappearance of names associated
with origin and use.

With the application of X-ray powder diffraction techniques to clay miner-
als it became obvious that these minerals fall into a few distinct groups and
Ross and Kerr (1931) gave three groups, namely, the kaolin group, the mont-
morillonite group, and the hydromica group and identified the major members
of these groups. In the 1930s the crystal structures of the phyllosilicates were
established (Pauling, 1930; Gruner, 1932, 1934; Hofmann, Endell and Wilm,
1933) and the idea of isomorphous substitution (Marshall, 1935; Hendricks,
1942) proposed as an explanation of cation-exchange capacity, and thence the
development has been rapid.

By 1939 clay mineralogy had so developed that Sedletskii gave an extremely
complex classification scheme for the colloidal minerals in soils which was
even further developed in a later publication (Sedletskii, 1945). In this
classification he gave 22 groups, in each of which was given at least three, and
frequently many more, distinct species divided into stabilites, mutabilites,
and metastabilites, stabilites being crystalline species, mutabilites gels of the
same composition and metastabilites something intermediate, considered as
being the materials formed as mutabilites crystallized into stabilites. In
addition, Sedletskii (1945) also had various groups of sesquioxides, humates,
humussilicates and carbonates, also divided under these three main headings.
Such a classification scheme, while an interesting intellectual exercise, is
unlikely to be of great practical value.

More recently there has been considerable agitation to devise an inter-
nationally acceptable classification scheme and in view of the fact that there
is still argument as to the best mode of classification to adopt, it is undesirable

TABLE 1.—COMPARISON OF THE CLASSIFICATION SYSTEMS OF
KIRWAN (1794) AND DANA (1894)

Kirwan	Dana
Muriatic Genus	*Serpentine and Talc Division*
Magnesia mixed with Silex (Silicimurite)	Serpentine (including antigorite, chrysotile, etc.)
Keffekill (myrsen, meerschaum)	Totaigite
Martial muriatic spar	Zöblitzite
Magnesia mixed with a notable proportion of calcareous earth and some iron	Metaxoite
Calcimurite	Hydrophite
Mild magnesia mixed with clay and iron	Aphrodite
Argillo-murite	Cerolite
Chlorite	Limbachite
In a loose form	Deweylite
Indurated and crystallized	Genthite
Slaty	Garnierite
Talc	De Saulesite
Talcite	Pimelite
Talc (common talc, Venetian talc)	Alipite
Shistose talc	Refdanskite
Steatites	Talc
Semi-indurated (craie de Briançon, Specstein)	Talcoid
Indurated steatites (lardites, Schmeerstein)	Sepiolite
Foliated or striated (talcites)	Connarite
Pot-stone (lapis ollaris, verharteter Talk or Topfstein)	Spadaite
Serpentine	Saponite
Asbestus (asbeste non mur)	Celadonite
Ligniform asbestus	Glauconite
Amianthus	Pholidolite
Suber montanum (corium montanum)	
Amianthinite (asbestartiger Strahlstein)	
Asbestinite (gemeiner Strahlstein)	
Asbestoid	
Agrillaceous Genus	*Kaolin Division*
Argillaceous Earths	Kaolinite
Native Argill (lac lunae, remethon Erde)	Meerschaluminite
Porcelain clay (kaolin)	Rectorite
Common clays in various states of induration	Leverrierite
Potter's clay	Halloysite
Indurated clay (verharteter Thon)	Newtonite
Shistose clay	Cimolite
Slate clay (shale, schiefer Thon)	Montmorillonite
Bituminous shale (Brandschiefer)	Razoumovskyn
Fuller's earth (Walkererde, smectis)	Pyrophyllite
Finer clays in various states of induration	Gümbelite
Lithomarga (Steinmark)	Neurolite
	Biharite
	Allophane

TABLE 1 (continued)

Friable or crumbling kind	Carolathine
Indurated lithomarga	Samoite
Bole	Collyrite
Argillaceous marls	Dillnite
Calorific earths, or those which	Schrötterite
strongly stain the fingers	Scarbroite
Red (reddle, rubrica fabrilis, Rothel)	
Yellow earth (gelbe Erde)	*Earthy Aluminous Silicates of Doubtful*
Black earth (black chalk, schwartze	*Character*
Kreide, zeichen Schiefer, pierre	Sinopite, melinite, ochran, plinthite,
noir)	smectite, rhodalite, sphragidite, eh-
Green earth	renbergite, portite, teratolite, catli-
Umber	nite, keffekilite, oravitzite, hverlera,
Harsh and rough earths	wolchonskoite, miloschite, selwynite.
Tripoli	
Pouzzolana (terras)	

to enter into the controversy here. Suffice it to say that general schemes have been proposed by Grim (1953), Brindley (1955), Brown (1955), Caillère and Hénin (1957), Hosking (1957), Strunz (1957), Frank-Kamenetskii (1958, 1960), Mackenzie (1959), and Lazarenko (1959). Definite international adoption of an agreed scheme must be left for the future, but the author would like to stress that any scheme adopted ought to have the following attributes:

(a) it must be logical;
(b) it must be easily remembered;
(c) the names must not be too cumbersome to use, in speech or in writing;
(d) it must be applicable in practice;
(e) it ought to be acceptable semantically (Robertson, 1962).

Because of these factors, and particularly (d), it would seem that there is a scope for employment of short group names (on a par with feldspar, mica, zeolite, etc.) and the dropping of variety names in preference to adjectives or prefixes such as ferrian, nickelian, or 2 M, Tc, etc.

ORIGIN OF CLAYS AND CLAY MINERALS

Discussion of the definitions of clays and clay minerals naturally leads one on to consideration of their origin. These materials may be formed by either hydrothermal or epigene processes and consequently clay minerals may be regarded as secondary minerals. Indeed, Bottini (1945) has commented that:

"The products of decomposition can, to a good approximation, be identified with the clay materials."

However, definitions are again troublesome, since, for example, what is termed a primary mineral by the pedologist or sedimentary petrologist—in

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that it is part of the parent material of the soil or rock—may be a secondary mineral to the igneous petrologist, and we also have to take into account the fact that clay minerals can be found in apparently fresh igneous rocks (Smith, 1960). Again, therefore, the definition depends upon the discipline, but in this instance there is at least the possibility of a reasonable definition for each discipline.

Ancient ideas on the origin of earths are now largely of historical interest. Theophrastus (*ca.* 300 B.C.) comments that:

> "it would seem that all types of earth are produced by fire",

and later:

> "some seem to have been set on fire and burnt: ... to put it plainly all ... result from a dry and smoky exhalation",

although he also remarks that:

> "these substances ... are produced either by some conflux or by percolation".

The fact that clays can be formed by the weathering of hard rocks has, however, been known from relatively early times and in the earlier part of the 19th century much credence was given to the idea that rock decomposition could result from electric currents set up at the conjunction of different rock types (Fournet, 1833; Brongniart and Malaguti, 1841). The latter authors indeed carried out experiments on the decomposition of feldspar on electrolysis. The action of rain was mentioned by Agricola (1546), and Fournet (1833) also referred to the efficacy of water containing CO_2 in promoting the decomposition of igneous rocks.

Many large deposits of clays, e.g. the kaolin deposits of Cornwall, are of hydrothermal origin, but, while these may be of considerable commercial importance by far the most widespread are those formed by epigene, or largely epigene, agencies, covering as they do soils and sedimentary rocks.

Epigene processes causing degradation of rocks may be conveniently divided into two classes: (a) physical and (b) chemical. The former derives largely from the effect of heat and cold in shattering rocks and while it may not *per se* cause the development of clay minerals it provides smaller masses of rock which are then more readily altered. In soil formation one physical weathering process of great importance is the grinding action of ice which has produced large areas of ground moraine or till, but the possibility of a certain amount of chemical alteration under such conditions cannot be excluded since there are usually distinct differences in mineralogy between the parent rock and the till derived therefrom. It seems unlikely that these have been produced simply by percolating waters in the period succeeding the Ice Age, although this factor cannot be entirely neglected—and this view is supported by the fact that even short-term dry-grinding of minerals can produce very marked changes and alteration (e.g. Mackenzie, Meldau and Farmer, 1956).

Chemical weathering can again be subdivided into (a) atmospheric and (b) biological. The former includes the action of rain and water with their dissolved gases, which can lead to hydrolysis, carbonation and oxidation of

rock-forming minerals. In many geological texts one finds the weathering of feldspar represented as

$$2KAlSi_3O_8 + 2H_2O + CO_2 = Al_2Si_2O_5(OH)_4 + K_2CO_3 + 4SiO_2$$
orthoclase   kaolinite

Yet such a simple transformation is by no means universal, and in our experience illite appears to be perhaps more readily formed by the weathering of feldspar than does kaolinite. The fallacy of accepting this reaction as fundamental has been mentioned by several workers and Yaalon (1959) has reported that feldspar can give rise to kaolinite, illite or montmorillonite depending on the prevailing conditions. Such fundamental changes probably necessitate the inclusion of a solution or near-solution phase as the structural alterations in the mineral appear to be too drastic to occur by simple ion migration and hydration—although DeVore (1959) has suggested a relationship with the crystallographic direction of the weathering process in the feldspar crystal. Other transformations, however, seem definitely to occur by ion migration and perhaps reorientation of groups, since the products have a definite crystalline orientation with respect to the original materials (i.e. the transformation is topotactic), as is evidenced in the alteration of olivine crystals to iddingsite (Smith, 1961) or the less intense changes involved in the alteration of biotite to vermiculite (Walker, 1949).

Whether an alteration is topotactic or not may at times be difficult to decide, and this has led to much controversy in the past. The work of Hénin and collaborators (Hénin and Robichet, 1954; Caillère, Hénin and Esquevin, 1957, 1958) on dilute solutions has shown the facility with which crystalline clay minerals can be formed from solution, and Roberston (1961) has recently proposed a solution theory for the origin of English fuller's earths, based on close similarity of composition over a widespread area. It is clear, therefore, that in assessing the nature of the weathering or alteration process it is necessary to consider in detail not only the particular material in question and its parent material, but also the distribution of similar materials over a wide area and fundamental laboratory investigations which give an insight into possible reaction processes.

The contribution of biological agencies to clay mineral formation is still incompletely understood. It has been known or assumed for a considerable period that the organic material present in soils and derived from growing plants or micro-organisms (see Merrill, 1906) can give rise to quite considerable alteration of the minerals originally present. However, with the refined methods now available it is possible to determine more accurately the mechanism of the processes occurring. For example, although it has long been known that primary minerals can be decomposed by micro-organisms (e.g. Müntz, 1890; Merrill, 1906; Oberlies and Pohlmann, 1958; Oberlies, 1958), it is only recently that it has been shown by Duff, Webley and collaborators (Duff and Webley, 1959; Webley, Duff and Mitchell, 1960; Duff, Webley and Scott, 1962) who studied micro-organisms grown in pure culture, that the

active agent in certain instances is 2-ketogluconic acid (which is a product of the metabolism of these micro-organisms) and that amorphous material remains. A similar mechanism may operate with lichens and mosses (see Jacks, 1953), and it is suggestive that the amount of amorphous materials found in the clay fractions of certain soils is greatest in the upper horizons (Mitchell and Farmer, 1962) and that such amorphous material is richer in silica than is normal allophane. Such observations suggest that this material may have originated under the influence of the exudates from micro-organisms and plant roots. How far these amorphous materials can subsequently re-combine to form crystalline clay minerals is still a matter for conjecture and for further investigation, but undoubtedly the environment would here exert a critical influence.

The weathering of minerals in igneous or metamorphic rocks is, however, only part of the story, since sedimentary rocks are themselves weathered by the same processes. Here, physical weathering may be of importance on its own account, since it may cause disaggregation of the individual particles leading to material which is identical with that in the original rock. Under chemical weathering the amount of change occurring will obviously depend upon the stability of the minerals under the conditions in which weathering takes place, and upon the time factor involved. Given a sufficiently long period one would expect the same secondary minerals to be predominant in one specific environment provided the original parent rocks, be they igneous or sedimentary, were not dissimilar in chemical composition.

The problem of the alteration of minerals during transport and sedimenta-tion is one which is perhaps of more interest to the sedimentary petrologist than to the pedologist. It is closely connected with weathering, however, since one of the main problems is how far the clay mineralogy of a sediment is affected by its environment during transportation and sedimentation and how far it reflects the weathering conditions on the land surface from which it is originally derived. Millot (1949), from the examination of a considerable number of sedimentary clay rocks in France, concluded that the predominant clay mineral can be related to the conditions of sedimentation. Weaver (1959), on the other hand, shows that in many instances the mineralogy of sediments is related more closely to the mineralogy of the land surface. The same con-clusion may be drawn from the fact that the predominant clay mineral present in limestone, as reflected in the mineralogy of the soils derived there-from (Yaalon, 1955), may be kaolinite, illite, montmorillonite or palygorskite (Munoz Taboadela, 1953; Cecconi, 1953; Lippi-Boncambi, Mackenzie and Mitchell, 1955; Yaalon, 1955). It would appear, indeed, that one of the most important factors is the length of time during which land-derived materials are in contact with their sedimentation environment. In solving such problems laboratory experiments, such as those carried out by Whitehouse and McCarter (1958) on the effect of artificial sea water on specific clay minerals, have an important part to play.

In conclusion, the author would like to stress that field studies dealing with

clay formation will be incomplete without parallel laboratory investigations. In this instance one may refer to the classical work of the schools of Correns (Correns, 1961), Hénin (Caillère, Hénin, and Esquevin, 1957; Pedro, 1961), and Roy (Roy, 1961). Provided such studies are carried out under conditions simulating those believed to be operative in the field, the information obtained will be invaluable in the interpretation of natural processes.

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