CLAY MINERALS IN ARGILLACEOUS SEDIMENTS OF THE HIMALAYAN ZONE

BIBHUTI MUKHERJEE
Geological Survey of India, Calcutta, India

Received 20th December 1963

ABSTRACT: A study has been made of the relative abundance of clay minerals in about a hundred samples of sedimentary rocks of Pre-Cambrian, Cambrian, Upper Carboniferous, Permian, Triassic, Jurassic, Lower and Upper Cretaceous and Tertiary formations in the geosynclinal basin of the (Tethys) Himalayan zone. Chlorite predominates in the Cambrian sediments and illite in the Permo-Carboniferous to Tertiary sediments; kaolinite is a minor variable component and montmorillonite is scarce. The high-chlorite Cambrian sediments are rich in dolomite and gypsum and contain predominantly plagioclase felspars, whereas the high-illite Triassic and Eocene sediments are rich in alkali felspars, the dolomite is low and gypsum is absent. In the (newer) Tertiaries of the Middle Miocene age (fluviatile deposits in shallow freshwater basin) illite, kaolinite and montmorillonite are present in almost equal amounts but chlorite is undetectable. The predominance of kaolinite and montmorillonite in the sediments of the Lower Pleistocene period indicates the existence of a distinctly fluviatile facies in the basin of deposition. Interstratified illite-montmorillonite and montmorillonite are characteristic of Siwalik (Miocene) sediments and of Spiti-shale type Jurassic sediments.

INTRODUCTION

Millot (1949, 1952) studied argillaceous sediments from the Mesozoic and Tertiary rocks of France and concluded that (a) illite predominates in marine and lagoonal sediments with some kaolinite in the first but not in the second, (b) kaolinite is characteristic of the acid continental facies, and (c) illite is the principal clay mineral in the basic continental facies with montmorillonite as an occasional associate. Grim, Dietz & Bradley (1949) and Grim (1953) reported that montmorillonite is generally rare in ancient sediments of the United States, particularly those older than the Mesozoic; but that a few beds, mostly the Cretaceous formations of the Great Plains and the Gulf Coast, contain predominantly montmorillonite which is regarded as being of volcanic origin. In the Paleozoic shales of Illinois from Ordovician to Pennsylvanian formations Grim, Bradley & White (1957) found illite to be the most abundant component in the older Paleozoic beds and in the Pennsylvanian shales.
In these formations the relative abundance of chlorite and kaolinite varies widely and montmorillonite is very scarce. Weaver (1959) observed that illite predominates in the pre-Upper Mississippian sediments but is less abundant in the post-Lower Mississippian clay sites where montmorillonite and kaolinite are more common. In the Gulf Coast Eocene shales, Burst (1959) reported that illite is most abundant in the deltaic sandstones and chlorite in the beach sandstones; moreover, more chlorite is found in the deltaic, lagoonal and marine sandstones than in the adjacent shales.

Taylor (1952) suggested that, if diagenetic changes are disregarded, kaolinite sediments are likely to be formed under conditions of peneplanation and illitic sediments under those of the geosynclinal stage. According to Grim et al. (1949) marine diagenesis favours the transformation of kaolinite to illite, and consequently kaolinite-rich sediments should be of non-marine origin. Ross (1943) considered that kaolinite formation is associated with prolonged weathering and leaching, and montmorillonite with restricted oxidation and poor drainage. The importance of metamorphic processes in altering montmorillonite to mica-type minerals was stressed by Grim (1953); he also attributed the low kaolinite content of ancient sediments to the same processes. In view of the fact that in older Paleozoic sediments illites are of the 2M type and that 1M illites are generally rare in ancient sediments, Weaver (1959) considered that the 1M structure is altered to the 2M structure during the process of metamorphism.

**EXPERIMENTAL**

Since X-ray powder diffraction photographs of sedimentary rocks from Cambrian and Permo-Carboniferous formations showed practically no clay mineral patterns, the clay fraction (< 2 μ) of each sample was separated by sedimentation and dried at 105°C. For X-ray examination Fe Kα-radiation from a Norelco Philips stabilized X-ray unit operating at 38 kV, 10 mA, a calibrated powder camera (114.59 mm diameter) with a special slit system, and 30 hr exposures were used with samples uniformly packed into Lindemann glass capillary tubes of 0.3 mm diameter. Powder photographs of each sample were taken before and after glycerol treatment as well as after heat treatment at 520°C for 1 hr. Diffraction patterns of artificial mixtures of illite, kaolinite, montmorillonite and chlorite were obtained using aluminium powder as an internal standard for calibration of integrated intensities. The intensities of selected reflections of each component were measured with a Moll recording microphotometer. The area of the peak, above an estimated background level, was corrected using a log-sector calibration curve.
The relative amounts of the four components were evaluated from the integrated intensity ratios of the basal reflections, and the intensity contribution for the same basal reflection due to two components was assessed empirically from the intensity ratios of other reflections. Comparison of the intensity of the combined basal reflections of montmorillonite and chlorite and that of montmorillonite in glycerol-treated and heat-treated samples with the intensity of the original basal reflection of illite is rather a crude procedure for estimating montmorillonite, but at higher concentration the intensities of the 310 and 260 reflections of montmorillonite substantiated the determined values. The 003 and 005 reflections of chlorite, the 002 and 003 reflections of illite and the 003, 202 and 204 reflections of kaolinite were used for evaluation, and gave reasonable accuracy for a four-component mixture. Percentages of each component are given to the nearest 10% since differences between the relatively highly crystalline standards and the widely varying crystallinity of the samples made evaluation difficult. The 2M structure of the illite was confirmed in illite-rich sediments from the 025, 115 and 116 reflections. The trioctahedral nature of the chlorite in the Cambrian sediments was ascertained from the relative intensities of the sequence of basal reflections and from the 060 spacing. Interstratified illite-montmorillonite was considered to be present in montmorillonite-rich sediments because of the broad basal spacing at 10.6–11.0 Å, but this phase was not considered in quantitative assessment of the four-component system. Other poorly crystalline phases also occurred. The form-factor functions of scattering distribution within the angular range corresponding to \( \sin \theta/\lambda \) values of 0.16–0.14, 0.11–0.10 and 0.07–0.035 are not the same for all the minerals concerned. The integrated intensities of individual reflections of each component in the above range of \( \sin \theta/\lambda \) values were not compared with the intensity value of the 003 reflection of illite (Johns, Grim & Bradley, 1954) as the intensity contribution at 3.34 Å is uncertain because of the quartz reflection. The integrated intensity ratios of different reflections were, however, compared empirically for evaluation.

The accuracy of determination in the standards was estimated at about ±25% of each component for higher concentrations and about ±50% of each component for lower concentrations (the limit being set by the background uncertainty). The errors in the determinations result from many variable factors. Thus, the clay fractions of samples from different types of formation are not uniform in particle-size distribution and the crystallinity and the chemical composition of each component are not identical. Differential absorption effects due to the differences of matrix composition and crystallinity, and differences in the form-factor functions controlling peak asymmetry within the angular
The width of diffuse bands were not properly considered. The oriented aggregate technique was not used as the natural variation in the degree of preferred orientation and the non-uniform intensity distribution of the basal and other reflections limit the scope of comparative evaluation of integrated intensities (Norrish & Taylor, 1962). The method of Johns et al. (1954) for the calculation of intensity contribution to the strong 3.5 Å reflection due to the 004 peak of chlorite and the 002 of kaolinite, and subsequent corrections for montmorillonite after ethylene-glycol treatment, and for kaolinite after heat treatment at 450°C, is not quite satisfactory as the spacings for chlorite and kaolinite vary in the range 3.52–3.59 Å resulting in a double peak or a broad peak; moreover, the 005 reflection of untreated montmorillonite is very weak even at higher concentration.

RESULTS

The sediments investigated occur as beds of enormous thickness and were laid down in a vast geosyncline continuously from the Cambrian to early Tertiary (Krishnan, 1960; Wadia, 1961). About a hundred samples of sedimentary rocks from these beds were obtained from the collection made during detailed stratigraphical and palaeontological studies to determine the age of the formations. The locality of origin and a description of each sample are recorded in the register of the Indian Museum. Complete environmental data, sedimentation conditions and epigenetic alteration are undeterminable factors for the samples studied, but some information about the type of formation and its origin are available from the old records (Gee, 1934, 1947; Middlemiss, 1890, 1891, 1896; Griesbach, 1891, 1893; Stoliczka, 1865; Wynne, 1877, 1878; Hayden, 1904; Pilgrim, 1910; Auden, 1934). In addition to the clay minerals the non-clay components in each sample were also studied, and the alkali felspars and plagioclases were determined by the procedures of Goodyear & Duffin (1954) and Donnay & Donnay (1952), and confirmed by optical study.

Cambrian sediments

Marine fossiliferous rocks of Cambrian age are prominent in a thick series of strata in the geosynclinal basin of the (Tethys) Himalayan zone. A thick sequence of Purple Sandstones of the Cambrian formation is well developed in the southern part of the Khasor Range to the north-west of Saiduwali and also in different parts of the Salt Range. The Purple Sandstones, so named because of their colour, are considered to be marine shallow-water deposits of a semi-arid climate. Six samples from the Khasor Range near Saiduwali and from the Salt Range near Chel Mt. Ridge, Basharat and Nara contained 60–70% chlorite, 30–40% illite, 0–10% kaolinite; montmorillonite was not detected. The
Neobolus beds, also of Cambrian age, are characterized by groups of dark micaceous shales, sandstones and conglomerates with white dolomitic layers, and the six samples examined from the Khasor Range near Saiduwali and from the Eastern Salt Range near Jalalpur contained 60–80% chlorite, 20–30% illite, 0–10% kaolinite; montmorillonite again was not detected. The non-clay accessory minerals were mainly gypsum, dolomite, calcite, felspars (predominantly plagioclase), hematite and quartz. The high-chlorite Cambrian sediments are rich in gypsum and dolomite with plagioclase felspars as a subordinate component.

**Upper Carboniferous sediments**

Following the Cambrian salt-pseudomorph shales the marine facies reappeared with the deposition of the Upper Carboniferous sediments which are perfectly developed in the Salt Range. The Speckled Sandstones are brownish sandstones with a mottled or speckled appearance; they are current-bedded and show evidence of shallow-water deposition. The ‘chert in lavender clay stage’ consists of lavender-, purple-, and grey-coloured shaly rocks and associated sandstones of the Speckled Sandstone series; again current bedding and ripple marks indicate shallow-water deposition. Four samples of Speckled Sandstone and Lavender Clay-Chert from near Makrach, Sanwans and Ghundi in the Salt Range show the presence of 40–50% illite, 40–50% chlorite and 10–20% kaolinite with no detectable amount of montmorillonite. The non-clay accessories are mainly dolomite, calcite, hematite, felspars, quartz, and often small amounts of gypsum.

**Permian, Triassic and Jurassic sediments**

The basin of deposition after Upper Carboniferous times is thought to have been deeper during the Lower Permian period. The Permian beds are characterized by sandy calcareous material indicating shallow-water deposition. The Productus Limestones of the Lower, Middle and Upper Permian beds are perfectly developed in the Salt Range, and six samples of Productus Shaly Limestone from near Nilawan, Jalhar, Sathra and Golewali contained 50–60% chlorite, 30–40% illite, 0–10% kaolinite and no detectable amount of montmorillonite. The non-clay accessories are mainly gypsum, calcite, dolomite, hematite and quartz.

In the Simla Himalayas a thick series of carbonaceous dark slaty shales with brown quartzite partings referred to as the Infra-Krol Series has been provisionally correlated with the Permian. The succeeding series of thick orange-brown sandstones and shales, known as Krol Series, shows signs of shallow-water deposition and is believed to be homotaxial with the Sirban Limestone of Hazara and the Productus Limestone of the Salt Range (Auden, 1934). The Krol Shales with
ripple marks are well developed in the Krol belt of Sirmar State. The four samples of Infra-Krol Shale and Krol Red Shale from the type localities near Solan and Kando in Sirmar State consisted of 60–70% illite, 20–30% chlorite, 0–20% kaolinite and 0–10% montmorillonite. The non-clay accessories are mainly felspars (predominantly alkali felspars), calcite, hematite, quartz and small amounts of dolomite. The high-illite Krol Shales are rich in alkali felspars and poor in dolomite; gypsum is absent.

Marine Triassic deposits of shallow-water origin are more or less completely developed in the Salt Range. Eight samples of sandstone, shaly limestone and conglomerate from the Triassic beds of the Salt Range near Sarai, Amb and Nammal, and from the Khasor Range below Kingriali Ridge had the composition 40–60% illite, 20–40% chlorite and 20–30% kaolinite with no detectable montmorillonite. The non-clay accessories are mainly calcite, dolomite, felspars, hematite, quartz, and often gypsum and goethite. The high-illite and low-chlorite samples contain predominantly alkali felspars, plagioclases being more abundant than alkali felspars in some chlorite-rich samples.

Jurassic strata of considerable thickness consisting of limestones, sandstones and shales overlie the Triassic stages. In the Salt Range proper the Variegated Series of Jurassic beds are considered to have followed a short period of sub-aerial conditions. Six samples of Jurassic Variegated Sandstones from near Pai Khel, Buri Khel and Sakesar Ridge in the Salt Range contained 40–50% illite, 30–40% kaolinite, 0–20% chlorite and 0–10% montmorillonite. The non-clay accessories are mainly felspars (predominantly alkali felspars), quartz, hematite, calcite, dolomite (minor), and often goethite.

The Hazara sequence consisting of Permo-Carboniferous to Jurassic sediments is completely developed in a number of sections in the Sirban mountain near Abbottabad, Hazara; features of the north-western zone are massive limestones overlain by the typical Spiti Shales. Six samples of Spiti Shales consisted of 50–60% illite, 20–30% montmorillonite, 10–20% kaolinite and no detectable amount of chlorite. Interstratified illite-montmorillonite is present in all samples. The non-clay accessories are mainly felspars (predominantly alkali felspars), calcite, quartz, hematite and dolomite.

**Cretaceous sediments**

The calcareous and argillaceous rocks of the Cretaceous system are perfectly developed in Baluchistan as a series of limestones and shales in the eastern zone and as dark greenish grey sandstones and sandy shales in the western zone. Lower Cretaceous beds of shallow-water formations occur in the western Salt Range and in the Surgarh Range as a
Clay minerals of the Himalayan zone

series of shales (containing abundant belemnites) and sandstones. Ten samples of sandstone, conglomerate and shale from Baluchistan (near Kaisergarh, Takht-i-Suliman Range), from south Afghanistan (near Kokoran, Argandab and Killa Abdullah) and from the Surgarh Range (near Malla Khel) were examined. They contained 30–50% illite, 20–50% chlorite, 20–30% kaolinite and 0–10% montmorillonite. The Upper Cretaceous sequence is completely developed on a large scale in Baluchistan and extends over wide areas in south Afghanistan. Six samples of sandstone, conglomerate and shale from Baluchistan (near Dozan, Bolan Pass and Kari, Quetta) and from south Afghanistan (near Naoroji, Argandab) consisted of 40–50% illite, 20–30% chlorite, 20–30% kaolinite and 0–10% montmorillonite. The non-clay accessories are mainly calcite, feldspars, quartz, hematite, dolomite, and often goethite and gypsum (minor).

Tertiary sediments

Tertiary sediments are quite extensive in Sind, Baluchistan, Afghanistan, the Trans-Indus Ranges and the north-west Himalayas, and in all these areas a lower marine facies and an upper fresh-water or sub-aerial facies are recognized. In Baluchistan the calcareous and argillaceous sediments of Mesozoic and Eocene ages are of marine origin and are followed by Upper Tertiary sediments of brackish- to fresh-water origin.

The Ranges of the Sind-Baluchistan border show an excellent development of sediments of Ranikot stage (Lower Eocene) formed under extensive marine conditions, and a thick band of shaly limestone containing Nummulites is developed typically in the Bolan Pass. Six samples of shale of the Ranikot series from Baluchistan (near Much, Bolan Pass) and from south Afghanistan (near Ghaziaband Pass), and two samples of Nummulitic Shale (Middle Eocene) from Baluchistan (near Kirtha, Bolan Pass) had the composition 50–70% illite, 20–30% kaolinite, 0–20% chlorite and 0–20% montmorillonite. Marine Tertiary sediments are developed in the Upper Indus Valley in Ladakh along a long narrow zone parallel to the Himalayan axis from Kargil. They consist of calcareous shales overlain by thick bands of shaly limestone and sandstone. Four samples of shaly sandstone and calcareous shale of Eocene age from Ladakh (near Kargil and Pugu) consisted of 50–60% illite, 20–30% kaolinite, 0–10% chlorite and 0–10% montmorillonite. The non-clay accessories are mainly calcite, feldspars (predominantly alkali feldspars), quartz, hematite and rarely dolomite. The high-illite Eocene sediments are rich in alkali feldspars and poor in dolomite.

The Saline Series at Kalabagh in the north-western portion of the Salt Range is of uncertain age. It is considered by Pascoe (1921) and
Christie (1914) to consist of sedimentary salt beds of Tertiary age, but Gee (1934, 1947) believed that the parts of the Saline Series to the north-west of Kohat were of Eocene age and those in the Kalabagh area were of Cambrian age. Six samples of sandstone and shale from the Salt Range (near Kalabagh, Nilawan and Katha Masral) contained 40–50% illite, 20–40% chlorite, 10–30% kaolinite and 0–10% montmorillonite. The non-clay accessories are mainly dolomite, calcite, felspars, quartz and often gypsum. Four samples of typical red salt-marl of the Saline Series from near Kalabagh and Chittidil had the composition 60–70% chlorite, 20–30% illite, 0–10% kaolinite with no montmorillonite—a composition similar to that of the Cambrian sediments. The non-clay accessories are mainly gypsum, dolomite, quartz and hematite.

The newer Tertiaries are developed on a large scale in the extra-Peninsular region, where they form the low outermost hills of the Himalayas. The Siwalik strata which commenced to form in the Middle Miocene have all the characteristics of fluviatile deposits (alluvial detritus) laid down in a shallow fresh-water basin. In the type areas of Siwalik at Potwar, Kangra, and parts of the north-west Punjab a continuous sedimentation took place from the Ranikot stage to the Lower Pleistocene period with a gradual change from a brackish environment to increasingly fresh-water conditions for deposition, this was accompanied by a change from lacustrine to fluviatile conditions. Three samples of sandstone and Nummulitic Grit of the Kamlial stage of the Lower Siwalik formations in the Potwar area and in the Salt Range near Bhalwal contained 40–50% illite, 20–30% kaolinite, 20–30% montmorillonite and no chlorite was detected. Six samples of calcified sandstone and sandy-clay from the Siwalik formations of Dehra Dun, Potwar, and other areas of Middle Miocene age in the north-west Punjab consisted of 40–50% illite, 30–40% kaolinite and 20–30% montmorillonite with no detectable amount of chlorite. The composition of the Upper Siwalik deposit indicates a significant change in the character of the sediments, since six samples of 'sandy-clay in soft sandstone' from near Murree and Nara Chita Pahar consisted of 40–50% kaolinite, 30–40% montmorillonite and 20–30% illite with no detectable chlorite. Interstratified illite-montmorillonite is present in most of the samples of the Siwalik formations. The non-clay accessories are mainly calcite, felspars (predominantly alkali felspars), quartz, hematite and dolomite (scarce).

CONCLUSIONS
The marine shallow-water sediments of the Cambrian formations consist mainly of trioctahedral chlorite, and some illite with occasionally
Clay minerals of the Himalayan zone

kaolinite but without montmorillonite. In the Upper Carboniferous and Permian sediments both chlorite and illite are abundant, kaolinite is a subordinate component and montmorillonite is an infrequent associate. Illite (of the 2M type) is most abundant in marine sediments of the Triassic and Jurassic beds, chlorite and kaolinite are the usual subordinate components, and montmorillonite is an infrequent associate. Interstratified illite-montmorillonite occurs in the Jurassic Spiti Shales which consist mainly of illite and montmorillonite.

In Lower and Upper Cretaceous sediments deposited in shallow-water environments illite and chlorite are more abundant than kaolinite. The Tertiary sediments of Eocene age are characterized predominantly by illite; both kaolinite and chlorite may be present, although chlorite is often absent, and montmorillonite is rare. The complete succession of marine shallow-water deposits from Pre-Cambrian to Tertiary in different areas of the extra-Peninsula shows the predominance of chlorite in the Cambrian sediments and of illite in the Permo-Carboniferous to Tertiary sediments; kaolinite is a subordinate component varying within wide limits and montmorillonite is scarce. The high-chlorite Cambrian sediments are rich in dolomite and gypsum with predominantly plagioclase felspars, whereas the high-illite Triassic and Eocene sediments are rich in alkali felspars but poor in dolomite and gypsum is absent.

In the newer Tertiary deposits of Middle Miocene age (fluvial deposits in shallow fresh-water basins) illite, kaolinite and montmorillonite are present in almost equal amounts but chlorite is undetectable. The predominance of both kaolinite and montmorillonite over illite in the Upper Siwalik sediments is considered as indicating the existence of a distinctly fluvial facies in the basin of deposition. Interstratified illite-montmorillonite is usually present in the Siwalik sediments.

ACKNOWLEDGMENTS

The author is most grateful to Dr B. C. Roy, Director-General of the Geological Survey of India, for kind permission to work on registered samples of the Indian Museum and to publish this paper, and to Dr M. V. N. Murthy for providing facilities to select samples from the Indian Museum gallery and for his interest in the work.

REFERENCES

MILLOT G. (1949) Geol. appl. 2, Nos. 2-4, 352.