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SOME RESULTS OF THE STUDY OF CLAY MINERALS IN THE U.S.S.R.*

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Abstract—Clay mineral studies in the U.S.S.R., mainly since 1960, are summarized. Electron diffraction techniques especially have been developed and used extensively. Methods have been developed for simultaneous recording of DTA curves and other properties. Polytypism of phyllosilicates has been studied intensively by Zvyagin and others. Much new information on the structure and morphology of halloysites has been obtained by electron-optical methods. Other clay minerals extensively studied include allophanes, kaolinites, chlorites ("donbassite" is preferred to "sudoite" for dioctahedral varieties), alushtite (or tosudite), monothermite, and various zinc-bearing clays. Much attention has been given to physico-chemical and colloid-chemical properties. The natural occurrence and alteration of clays and clay minerals are described in weathering and oxidation zones, in soils, and in Recent and Ancient sediments. Primary clay minerals of hydrothermal origin are described.

INTRODUCTION

THE study of clay minerals in the U.S.S.R., as in many other countries, was begun in connection with the practical utilization of kaolinitic and montmorillonitic clays. At the end of the nineteen thirties and forties the interest in clay material had sharply increased; a great number of publications appeared on the mineral composition of clays, their origin and practical use. Concurrently, research began on the clay minerals in the weathering zone, the oxidation zone, soils and sedimentary rocks; much attention was paid to the role of clay minerals in engineering construction. Somewhat later there was a broad development of investigations devoted to the physico-chemical and colloid-chemical properties of clay minerals and to their crystal-chemistry.

The author's aim is to present some results of the study of clay material by Soviet specialists, mainly during the last decade.

METHODS OF RESEARCH

Along with the methods widely used in other countries, electron diffraction especially has been used for quite some time in the Soviet Union for the study of phyllosilicates. Electron diffraction methods have been developed further and their range and application expanded, particularly by Zvyagin (1967). Formulas have been derived and diffraction criteria established, which characterize disorder in phyllosilicates (Zvyagin, 1964). Some minerals have been studied with a 400–kV electron microscope electron diffraction unit, designed by N. M. Popov.

The latest electron microscope procedures for study of clay minerals include decoration (Gritsaenko and Samotoyin, 1966) and use of the moiré effect (Gritsaenko and Gorshkov, 1966). The method of electron micro-diffraction in electron microscopy is becoming a necessary auxiliary means for structure analysis of individual mineral particles, and for study of their internal morphology and microcrystallographic features (Shitov and Zvyagin, 1965). The possibility of an electron–microscopic study of the microstructure and microtexture of clayey and loess rocks (Il'inskaya, 1966) has been demonstrated.

The use of secondary diffraction effects seems quite promising. They offer the possibility of a quantitative characterization of regular combinations (epitaxial intergrowths) of the crystals in minerals, of a deciphering of the moiré patterns reflecting the specific features of the actual structure of minerals and of their defects (Zvyagin and Gorshkov, 1966).

Yu. S. D'yakonov (1962) has suggested a new method of direct deciphering of X-ray patterns of

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mixed-layer minerals by means of Fourier transformations. A method of computing optical constants has been worked out for various polytype varieties of phyllosilicates on the basis of an optical characteristic of some single representative of a given group of polytypes (Drits, Murav'ev and Shutov, 1966).

Great credit is due L. G. Berg (Kazan') and A. I. Tsvetkov (Moscow) for improvements in the equipment and theoretical foundations of the thermal analysis method. Thermal analysis has been widely used in our country since as early as the nineteen twenties. The DTA (differential thermal analysis) curves are recorded photographically. A recent feature in the use of this method is recording of DTA curves in a controlled atmosphere and the simultaneous recording of various heating curves, e.g. DTA and weight-loss curves; DTA, weight-loss and sample shrinkage curves; DTA and electroconductivity curves, etc.

Infrared spectroscopy produced most interesting results in a comparison of various vermiculites based on their oxonium content (Arkhipenko *et al.*, 1963); however, the degree of definition in the results obtained is not clear owing to the general complexity of the problem of oxonium in minerals.

STRUCTURAL STUDIES OF CLAY MINERALS AND ALLIED SILICATES

Polytypism

B. B. Zvyagin and co-authors have deduced theoretically and found experimentally unique polytypic varieties of chlorites (Zvyagin and Mishchenko, 1965), serpentines (Zvyagin, Mishchenko, and Shutov, 1966) and micas (Zvyagin, 1966). Based on the study of a large number of halloysite samples, the following deductions were made regarding the nature of this mineral (Zvyagin *et al.*, 1966; Chukhrov *et al.*, 1966; Chukhrov and Zvyagin, 1966).

Halloysites producing basal reflections of about 7 Å represent an independent kaolinitic polytype, the idealized structure of which is characterized by a double-layer repeat sequence (modification of variety 2 M_1). Actual structures are characterized by substantial disturbances in the alternation order of the layers and in the orderliness of their structure; from these features, it is possible to distinguish the halloysite series by X-rays and electron diffraction. Indirect indications of the extent of deviations of the structure of halloysite from the ideal model can be judged from thermal analysis data ($\tan\alpha/\tan\beta$ ratio and the shape index) and by i.r. spectroscopy (intensity ratio of absorption bands in the 3625–3695 cm^{-1} region). The degree of order in the halloysite structure increases as its content of weakly combined water diminishes.

Halloysite particles, when not subjected to redeposition, are polycrystals of prismatic habit consisting of several sectorial zones (platelets) with a common axis of elongation, which is also the axis of the prismatic crystals; this axis is usually near the crystallographic *b* axis, but sometimes approaches the *a* axis, or occupies an intermediate position. Crystallites with several directions of elongation are also encountered (Gorshkov, 1966). The faces of the prismatic crystallites are parallel to the *a* and *b* axes. The ends of some polycrystallites show platy cleavages. The formation of hollow tubular particles, that can be observed under an electron microscope only in suspended preparations, is a secondary process associated with dispersion of the halloysite; cores separate from the halloysite polycrystallites, which become hollow. The zonal structure characteristic of halloysite polycrystallites (frequently with clearances between the zones), which can often be observed in cross section, is very favorable for liberating the cores; apparently, such separation of the cores and envelopes (which become hollow tubes) occurs repeatedly.

The available data do not constitute proof of the concept of transformation of halloysite into kaolinite or vice-versa. The primary form of halloysitic material is hydrated halloysite (endellite).

In some cases, halloysites pass through a vitreous allophane stage retaining the spherical or similar shape of particles typical for many of them and distinguishable in an electron microscope. Such, for example, is the white friable porcellaneous hydrated halloysite from the tertiary deposits of Bashkiria, which produces an X-ray pattern and DTA curve typical of the mineral (Borovko, 1960), or the hydrated halloysite–allophane from Bazar-tyube in Turkmenia (see below).

Allophanes

The study of allophanes from various deposits was the aim of special research. In Soviet mineralogical literature typical allophanes include the vitreous isotropic, usually virtually X-ray amorphous minerals with a variable $Al_2O_3:SiO_2$ ratio; some of them display transitions into earthy forms.

The author and his colleagues (Chukhrov *et al.*, 1964; Chukhrov *et al.*, 1965) have studied vitreous allophanes with $Al_2O_3:SiO_2$ ratios ranging from 1·00:1·06 to 1·01:2·00. The vitreous mineral from Turkmenia, externally markedly different from halloysite, proved to be very similar to it not only by composition, but also by DTA curve, X-ray powder pattern and i.r. spectrum. Its composition corresponds to the empirical formula 0·29 (Ca, Zn)O · $1{\cdot}00Al_2O_3 \cdot 1{\cdot}99SiO_2 \cdot 1{\cdot}96H_2O^+ \cdot 4{\cdot}49H_2O$.

Electron microscopically the mineral consists mainly of rounded particles; some single elongated particles are also observed. The other allophanes studied do not yield definite X-ray powder patterns; by DTA curves they do not differ essentially from other allophanes described in the literature and by i.r. spectra they exhibit a certain similarity to andalusite and kyanite. The presence on the allophane i.r. spectra of a number of variable absorption bands in the hydroxyl region (3500–3750 cm^{-1}), is a characteristic which may be attributed to hydroxyls of varying bond strength; their positions were established from thicker than usual preparations on vaseline-coated slides; the curves were recorded at a slow rate of scan. These data enable us to regard the typical allophanes as aluminum silicates hydrolyzed to varying degree and virtually amorphous to X-rays. The (OH) group in allophanes, in accordance with their amorphous nature, is bound far less strongly than in phyllosilicates. The alumina/silica ratio in allophanes can be said to depend upon the degree of hydrolysis of the amorphous freshly precipitated aluminum silicate. Studies of so-called phosphate allophanes from Kazakhstan and Czechoslovakia (Chukhrov *et al.*, 1965) showed that these forms cannot be regarded as mixtures of allophane and evansite, which makes the name allophane-evansite superfluous. There is more justification for considering the phosphate allophanes as fine gel mixtures of two X-ray amorphous compounds – of phosphate and aluminum silicate of variable composition, dependent upon the degree of hydrolysis.

Kaolinites

On the basis of an electron diffraction study, M. F. Vikulova and B. B. Zvyagin (1965) have conditionally deduced nine structural groups of kaolinite from the shape of the unit cell and degree of structural perfection. Unit cells with triclinic, triclino-pseudomonoclinic, and pseudomonoclinic symmetry are distinguished and, for each of these, three degrees of structural perfection are deduced. Kaolinites in protractedly formed weathering zones and authigenic kaolinites of sedimentary rocks have the most nearly perfect structures. Mechanical effects during redeposition promote the transformation of kaolinite of perfect structure and triclinic unit cell into a kaolinite of imperfect structure and psuedomonoclinic unit cell.

Chlorites

In 1906, Professor Ya. V. Samoĭlov described a mineral from Nagol'ny Ridge ore, which he classed with the members of the chlorite group richest in aluminum (up to 48·16 per cent Al_2O_3 and up to 1·38 per cent Fe_2O_3). E. K. Lazarenko (1940) established the distribution of minerals of this type in ore deposits of the Donets Basin and proposed the common name donbassites for them. From their external appearance, some donbassites have been taken for pyrophyllite. In H. Strunz's mineralogical handbook the donbassites are assigned to the kaolinite group. However, as has been shown recently (Drits and Lazarenko, 1966; Vlasov and Drits, 1967), the donbassites are dioctahedral chlorites. In 1963 G. Müller proposed calling the dioctahedral chlorites sudoites. However, it might be more appropriate to replace the term sudoite by the term donbassite, inasmuch as it was introduced earlier and refers to a typical dioctahedral chlorite.

Gümbelite

A detailed study of the gümbelite that forms transverse fibrous streaks (from fractions of a mm to 2 cm) in the shungite shales of Karelia (Drits, Zvyagin, Tokmakov, 1966) led to the conclusion that this mineral is a muscovite mica with a 2 M_2 structure and not 2 M_1, as was previously thought. The gümbelite fibers consist of rods elongated along *a* axis and packed so that alternately they form a ±60° angle with the elongation direction of the fiber; the angle between rods is 120° and they lie parallel to (001).

Alushtites, tosudite

A study of alushtite gave interesting results. This name was introduced by A. E. Fersman for a white, greenish or light bluish colored clay material found in quartz veins which intersect Jurassic shale in the Crimea. Recent studies have indicated that materials designated alushtite in some cases consist of only one mineral, which V. A. Frank-Kamenetskiĭ and co-authors (1965) have called tosudite. Some "alushtites" proved to be mainly mixtures of dickite and hydromica, occasionally with admixture of montmorillonitic mineral. Tosudite is an ordered mixed-layer form, consisting of alternating montmorillonite and dioctahedral chlorite (sudoite).

A mineral very like tosudite has been found also in streaks traversing rocks of the coal-bearing series of the Karaganda coal basin (Drits and Shutov, 1966). The name "sangarite" is given to orderly mixed-layer intergrowths (chlorite-like and vermiculite-like layers) from the cement of Mesozoic sandstone of the Sangar region in the Vilyuĭ depression (Drits and Kossovskaya, 1963).

Monothermite

Monothermite, first described by D. S. Belyankin (1938), is well-known in the Soviet literature. A specific feature is the presence on the DTA curve of only one endothermal peak at 550–600° and the absence of an exothermal peak at 950–1000° characteristic of kaolinite. Monothermite differs from kaolinite also in having a higher birefringence and a greater content of alkaline and alkaline-earth metals. Technologically monothermite is characterized by high plasticity and good refractory properties. Monothermite was discovered first in the Chasovyarsk deposit in the Ukraine, but later was described as a constituent of some clays in the Southern Urals. Monothermite from the Chasovyarsk deposit contains 3·25–3·56 per cent R_2O (mainly K_2O) and 0·59–1·05 per cent $CaO+MgO$ (CaO predominates). Mean analytical data lead to the formula $0{\cdot}2RO \cdot Al_2O_3 \cdot 3SiO_2 \cdot 1{\cdot}5H_2O \cdot 0{\cdot}5aq$. Yu. A. Rus'ko (1961) concluded that this clay is a composite of kaolinite (37·11 per cent) and hydromica (55·66 per cent) with a small admixture of free oxides.

Leverrierite

Leverrierite from the coal deposits of the Donets Basin is a mixture of dickite, "mixed-layer" mica and carbonaceous matter (Logvinenko and Frank-Kemenetskiĭ, 1961).

Zincsilite

Using this designation, N. A. Smol'yaninova and co-authors (1960) described an aluminum-free sauconite from the oxidation zone of zinc ores associated with sphalerite-bearing skarns (Central Kazakhstan). Zincsilite forms pseudomorphs after diopside. By DTA curve and X-ray pattern zincsilite is similar to sauconite; it swells in glycerine. The lamellae of the mineral are up to 2 mm in size. A zinc-bearing serpentine (serpophyte) containing up to 7·24 per cent zinc oxide has been found in serpentinized dolomite of the Kurgashinkan lead-zinc deposit in Uzbekistan (Golovanov, 1960).

PHYSICO-CHEMICAL AND OTHER PROPERTIES OF CLAY MINERALS

Extensive studies have been made of the physico-chemical and colloid-chemical characteristics of bentonite (Merabishvili, 1962) and of palygorskite (Ovcharenko *et al.*, 1963), the hydrophilic nature of clay minerals, the dependence of the colloid-chemical properties upon structure, the control of these properties, and the physico-chemical mechanics of clay minerals (Serb-Serbina and Rebinder, 1958; Ovcharenko *et al.*, 1965; Ovcharenko, 1966; Kruglitskiĭ, 1966, 1967).

To characterize the stability of clay dispersions, the concept of a "stability coefficient" (ratio of the magnitude of a relatively fast elastic deformation to the concentration of the disperse phase) has been introduced. The main methods used for controlling coagulation structures in clay-water systems are: ion exchange, reaction with reagents, mixing of clay minerals of different structure, and ultrasonic treatment. A study of the salt stability of clay minerals led to the conclusion that this property can be controlled by these methods within wide limits. Thus, by ultrasonic treatment, the salt stability of montmorillonite can be increased by a factor of 2–2·5 times. It has been shown that, along with palygorskite (attapulgite), a high salt stability is characteristic of some natural mixtures of palygorskite with montmorillonite, and mixtures of highly disperse illite with kaolinite.

Results have been obtained showing how the changes in dispersion or aggregation which occur in clay dispersions under the influence of ultrasound depend upon both the nature of the clay mineral, and the manner of the ultrasonic treatment. Kruglitskiĭ (1967) has recorded the formation of a montmorillonite with an imperfect structure after processing palygorskite with ultrasound. Experiments have clarified the ability of clay earths to undergo thixotropic transformations during vibratory drilling to depths of 5–6 m (Gumensky, 1960). Suggestions have been made for an electrochemical strengthening of unstable clay rocks (Titkov *et al.*, 1964), for instance, in drilling boreholes. The essence of this process is that, under the action of a direct electric current with addition of electrolytes, new minerals (gibbsite, hydrogoethite, hydrohematite, allophanes, hisingerite, calcite, etc.) are formed in the pores of the clays, which sharply increase the stability of the clay rock. Other important work on the engineering properties of clays has been undertaken by Priklonskiĭ *et al.*, (1958) and Sergeev *et al.*, (1963).

NATURAL OCCURRENCE, FORMATION AND ALTERATION OF CLAY MINERALS

Weathering zone and oxidation zone

The development of Ancient weathering zones depended to a great extent upon tectonic conditions. Very favorable conditions for the formation of weathering zones existed on wide and flat uplifts separated by big faults (Razumova and Kheraskov, 1963) before the appearance there of a sedimentary cover.

The oldest weathering zone on granites is in Karelia, where, as a result of metamorphism, the weathering products have altered to micaceous shaly rocks. In the Paleozoic, two main epochs are distinguished for the formation of weather zones—the Lower Paleozoic and the Middle Paleozoic. In the U.S.S.R., the Mesozoic (up to 100 m) and Tertiary (up to 50 m) weathering zones are especially well preserved; the first of these has been studied in great detail (Petrov, 1967). In the Urals and the Ukraine, an intensive kaolinization occurred from the Upper Triassic to the Middle Jurassic. In the northern and middle Urals, a washout of kaolinitic weathering zones began in the middle of the Cretaceous period but in the southern Urals and the Ukraine, it began in the Tertiary period; deposits of redeposited kaolinite virtually free of quartz admixture are known. A special feature of some kaolinitic weathering zones on granites in the Ukraine and the Urals is the presence in the lower part of the weathering zones (above the disintegration zone) of a zone of hydromica (illite) which higher up gradually changes to kaolinite (Petrov, 1967). However, there is often a direct replacement of feldspars by kaolinites, and the hydromica zone is absent (Kukovskiĭ, 1963). The weathering of a granite in regions where the loss of bases was moderate, gave rise not only to minerals of the kaolinite group, but also to montmorillonites (Chukhrov, 1960).

During Mesozoic weathering of serpentinites in the Urals, the products of which have been studied in great detail, a leached zone of "kerolith" (a mixture of serpentine and talc-like minerals) deweylite, trioctahedral montmorillonite and nickel silicates was formed above the disintegration zone of rocks containing small amounts of "kerolith"; above this lies the nontronite zone; the uppermost zone in the serpentinite–weathering profile is a zone of ochrous iron oxides formed after the nontronite (Rukavishnikova, 1958).

Supposedly, the Upper Triassic weathering zone formed on kimberlites of the diamond-bearing pipes of Yakutia and is characterized by a predominance of montmorillonite among the clay minerals (Rozhkov, 1963). Some old weathering zones occur under a cover of sedimentary rocks. For example, in the Angren coal-kaolin deposits, the kaolinitic weathering zone on Mesozoic and Paleozoic rocks is overlain by Jurassic sedimentary rocks (Petrov and Rubanov, 1960). About ten years ago, at a number of points on the Kola peninsula, a relict preglacial weathering zone overlain by morainic deposits was found resting on Precambrian rocks; the main mass of weathering products has been destroyed here by repeated glacial scouring (Afanass'ev, 1966). Of particular interest is the so-called sungulite (kol'skite) weathering zone developed on olivinites containing phlogopite. In the lower parts of the sungulite weathering zone, hydrophlogopite (mixed-layer forms of phlogopite and vermiculite) and vermiculite (after phlogopite) often occur. Still higher, vermiculite and sungulite (kol'skite) – a white, thin-flaky serpentine mineral, the formation of which has the character of vermiculite replacement (Petrov and Tokmakov, 1965) are found.

Available data indicate that the mineral composition of the ancient weathering products of silicate rocks has depended mainly upon their chemical composition and the climate. The formation of montmorillonitic minerals was facilitated by a higher content of bases in the original rocks or by their slow removal; unlike kaolinite, halloysite was forming under conditions of much more rapid weathering processes (Chukhrov *et al.*, 1966); the new data do not confirm the assertion that there is a difference in pH of formation of these two minerals as has been stated in the literature. Allophane formation was facilitated by a rapid interaction of sufficiently concentrated silica and alumina solutions with the formation of colloidal X-ray amorphous precipitates (Chukhrov *et al.*, 1964). Allophanes occur in regions where the products of Ancient weathering were destroyed by erosion after the Alpine orogenesis (Caucasus, Altai, Dzhungarian, Alatau, etc.) or by glacial denudation (Moscow region, Aldan, Kola peninsula, etc.). Since crystallized minerals, the composition of which would correspond to typical allophanes, have not been found in old weathering zones, it may be assumed that in the course of time allophanes, as very unstable minerals, become dissolved and cease to exist (geological ephemera); some allophanes alter to halloysite.

Present-day peculiarities in the mineralogy of the old weathering zones are determined more or less by processes taking place after their formation. One of these processes is opalization of the clay minerals. Over the territory of the Ukraine there is a wide distribution of essentially opaline rocks – pelicanites – formed by opal replacement of the more or less kaolinized rocks. Analogous rocks are found in Kazakhstan. The time when the pelicanites of the Ukraine were formed is not clear; in Kazakhstan they are overlain locally by Eocene sandstone and may be regarded as pre-Eocene (Ginzburg and Rukavishnikova, 1952). Finds of opalized bone fragments of a brontotherid (Eocene) in one of the sink holes of Central Kazakhstan indicates possible opalization also in post-Eocene times. In addition to the pelicanites, on the vast territory of Kazakhstan, opalization of clay mineral formations has occurred with preservation of their habits; fine mixtures of halloysite, kaolinite, and montmorillonite minerals with opal have developed; the chloropals – intensely opalized nontronites – are also included with formations of this kind (Chukhrov, 1962).

Soils. Considerable attention is given to the study of vermiculite, which has been established in a clayey fraction (less than 0·001 mm) of a sub-podzolized soil on granitic eluvium and on biotite-muscovite shale in Eastern Sayany, as well as in a turf-podzolic soil on a cover loam near the

Golitsino railway station (Moscow region). It was concluded that in soils on an eluvium of igneous and metamorphic rocks vermiculite is a new form, whereas, in soils on sedimentary rocks it is inherited (Gradusov *et al.*, 1966). Vermiculite from the Kovdorsk deposit (Kola peninsula) is used as a mulching material, as well as in hydroponics, as a filler for liquid fertilizers and chemical poisons (Gorbunov *et al.*, 1964).

Ordered mixed-layer forms are found in the soils of the U.S.S.R.: chlorite-vermiculite, hydrobiotite, rectorite (allevardite). The latter has been established in the less than 0·001 mm fraction extracted from soils on the micaceous shales of Karelia (Gradusov, 1967). The relation between the main types of soil formation and character of the finely disperse minerals in soils has been investigated (Gorbunov, 1966).

Recent and ancient sediments. Many publications are devoted to the characteristics of clay minerals in sedimentary rocks; some of them contain data on clay minerals in Recent sediments. Interesting generalizations have been made by M. A. Rateev (1964). Data on clay minerals in Recent basins of *humid zones* indicate no connection between the nature of clay minerals and the salinity of the sea water. In cores of marine sediments 10 m long from the surface of the sea floor, the clay minerals do not show any diagenetic alterations.

In the basins of the humid tropical zone that existed on the Russian platform in the geologic past, a change in the assemblages of clay minerals depended to a great extent upon the changes in shape and size of the basin, as well as upon the specific features of the supply areas. The special features of the mineral composition of the redeposited products of weathering for different climatic zones in which sedimentation is occurring are presented in Table 1.

Table 1. Clay minerals in the basins of humid zones

Climatic zones	Minerals
Tropical	Predominance of kaolinite, occurrence of halloysite and hydromica
Subtropical	Smaller amounts of kaolinite and greater amount of hydromica; montmorillonite appears; occurrence of halloysite
Cool	Predominance of hydromicas and montmorillonite. Kaolinite disappears or is very rare
Arctic	Hydromicas and chlorites are most abundant; occurrence of vermiculite; montmorillonite disappears

This climatic zoning in the distribution of clay minerals in Recent sediments corresponds to that found in soils. Authigenic mineral formation in the seas of the humid zone can result in the formation of montmorillonite from pyroclastic rocks of basic composition, the regeneration of micas, etc.

From the example of the basins that existed on the Russian platform in the geologic past, it may be concluded that in the marine basins of the *arid zones* the processes of authigenic mineral formation were proceeding more intensively and included the formation of palygorskite from montmorillonite or a trioctahedral mica, as well as the formation of sepiolite by chemical reactions. The latter forms localized lenses or thin beds; it is unknown as an allothigenous mineral.

The transformations of clay minerals in sediments by diagenesis, epigenesis (katagenesis), and metagenesis are discussed in numerous papers.

The alteration of biotite and muscovite in sedimentary rocks has been studied in detail by Kossovskaya *et al.* (1965); this process is a stage in the development of a number of mixed-layer forms (chlorite, vermiculite, dioctahedral montmorillonite); kaolinite is the final product. In contrast with biotite, muscovite in sedimentary rocks is more stable. On decomposition, according to A. G. Kossovskaya, usually no intermediate phases are formed and muscovite converts directly to kaolinite. Only a very finely dispersed muscovite can yield vermiculite-like minerals. So-called illites or hydromicas (degraded micas), so common in sedimentary rocks, can be products of a stepwise alteration of biotites, or originate during potassium fixation by montmorillonites; together with the transformed illites, the clastic illites can play a substantial role in sedimentary rocks. The above-characterized transformations are mainly typical of diagenesis.

In the zone of deep epigenesis and in the zone of metagenesis (transition zone to the zone of regional metamorphism) phyllosilicates of sedimentary rocks undergo a series of transformations (Shutov *et al.*, 1966). On platforms, these processes begin with burial of the sediments to a depth of 1·5–3·5 km. It has been noted that kaolinite alters to dickite and nacrite; hydromicas (illites) and chlorites are formed. In these zones a substantial role in the transformation and formation of clay minerals must be played by heated meteoric solutions, which explains the formation of dickite and nacrite in sedimentary rocks and in ore veins. Often, newly formed dickite in sedimentary rocks forms streaks, sometimes together with quartz. The same mode of origin is attributed to a mixed-layer mineral of the sudoite group, which together with dickite, nacrite, and carbonates forms streaks in the rocks of the coal-bearing series of Karaganda (Drits and Shutov, 1966).

G. V. Karpova (1966) has established that processes of post-sedimentational paragonitization are widely manifested in the different terrigenous Carboniferrous rocks of the Donets Basin. The formation of paragonite, at the stage of epigenesis and metagenesis, was favored by the composition of the rocks and by the action of the deep mineralized chloride waters.

Clay deposits in sedimentary strata. Among clays associated with sedimentary strata, a distinction should be made between clays formed *in situ* and redeposited clays.

Bentonite clays of many deposits are of an authigenic nature; the concept that their formation is the result of weathering of volcanic ashes and other volcanic products is widely held (Dzotsenidze and Machabeli, 1965). Most bentonites are of the alkaline-earth varieties.

In various regions of the U.S.S.R. there are also deposits of bentonite clays formed by redeposition of montmorillonite and other minerals in the weathering zone of effusive or metamorphic rocks. The best known of them is the Cherkassk deposit in the Ukraine, the clays of which were deposited in a shallow Lower Miocene basin. A particular feature of this deposit is the presence of a bed of palygorskitic (attapulgitic) clays up to 2 m thick in the middle of the bentonite mass (Ovcharenko *et al.*, 1966).

Refractory clays in the U.S.S.R. were formed mainly by redeposition of clay minerals of the weathering zone; they are associated chiefly with Carboniferous, Jurassic, Cretaceous and all Cenozoic deposits. Besides kaolinite and micas such clays contain montmorillonite, halloysite, and other minerals. The presence of halloysite is characteristic of the kaolin clays of the Troshkovsk deposit (Irkutsk area), which are associated with Jurassic deposits (Kychakov, 1958).

PRIMARY CLAY MINERALS OF HYDROTHERMAL DEPOSITS

Recent research has shown that dickite is the most common hydrothermal clay mineral in deposits of various types; in many cases, this mineral has been previously described as nacrite. True nacrite is very rare. Other hydrothermal clay minerals are donbassite, rectorite (allevardite) and tosudite. Table 3 lists examples of the occurrence of clay minerals in deposits of the U.S.S.R.

Available data on nacrite and dickite in hydrothermal deposits indicate that they were formed during late stages of mineralization later than vein quartz and ore minerals; the presence of dickite both in ore veins and in rocks, where there are no ore bodies of any kind (Crimea), indicates the absence of a direct relation between the formation of dickite and of the ore bodies. The late minerals of hydrothermal deposits include also donbassite, allevardite, and tosudite. There is, apparently, no substantial difference in the formation of dickite and of other hydrothermal clay minerals in ore

Table 2. Bentonite clays in certain deposits of the U.S.S.R.

Name of bentonite clay	Deposit	Age of beds	Notes
Gumbrin	Gumbri in Georgia	Senoman	—
Askan clay	Askana in Georgia	Eocene	Alkaline-earth bentonite, forms the top horizons of the deposit. Product of an acid activation of askan clay-askanite
Askangel'	Askana in Georgia	Eocene	Alkaline bentonite, forms lower horizons of the deposit
Askankol	Askana in Georgia	Eocene	Colloid fraction of askangel'
Oglan clay	Oglanly in Turkmenia	Paleogene	Alkaline-earth bentonite
Oglangel' (oglanlite)	Oglanly in Turkmenia	Paleogene	Alkaline bentonite
Nal'chikin	Nal'chik in Northern Caucasus	Tertiary	—
Kil (or Keffekelite)	Deposits of the Crimea	Cretaceous	Product of acid activation—krymsil'
Gil'abi	Deposits of Azerbaidzhan	Upper cretaceous—tertiary	Activated cretaceous bentonite from Khanlarsk deposit—Khanlarite

Table 3. Examples of occurrence of hydrothermal clay minerals

Name of deposit	Type of mineral bodies, paragenesis of minerals	Authors
Nikitovka mercury deposit (Donets Basin)	Dickite, quartz, cinnabar, pyrite and antimonite in fissures of shales, conglomerates and sandstones	Samoĭlov (1906) Logvinenko and Frank-Kamenetskiĭ (1961)
Tibsk mercury deposits (North Caucasus)	Quartz veins with cinnabar, calcite, dickite, and hydromica	Demidova (1959)
Lead-zinc deposits of Nagol'ny ridge (Donets Basin)	Quartz veins in sandstones, clay and sandy-clay shales with dickite, sphalerite, ankerite, donbassite	Samoĭlov (1906) Lazarenko (1940, 1950)
Stanitsa Tatsinskaya (Donets Basin)	Quartz-dickitic streaks (in sandstones) with calcite and rare segregations of galena and chalcopyrite	Zaritskiĭ (1960)
Crimea	Quartz-calcitic veins with dickite in an albite diabase, quartz-dickitic and dickitic veins in Tauric flysch	Kovalev (1947); Logvinenko and Frank-Kamenetskiĭ (1961, 1966)
East-Kounrad molybdenum deposit	High-temperature quartz veins (in granites) with dickite in cavities and in late transverse streaks	Chukhrov (1960)
Aktash deposit of alumina raw material (Uzbakistan)	Hydrothermally altered (in the crushing zones) acidic and intermediate effusives with tiny flakes and continuous masses of dickite	Zubrilina (1964)
Dashkensansk iron-ore deposit in Azerbaidzhan	Contact-metasomatic bodies; dickite forms segregations in volcanogene rocks or forms veins, streaks and pockets in alunitized and pyrophyllitized rocks	Kashkaĭ
Secondary quartzites of Central Kazakhstan	Metasomatic rocks (after various effusives) with quartz, dickite, alunite, andalusite, pyrite, rutile, diaspore	Nakovnik (1940)
Secondary quartzites of Beregovo foot-hills in Transcarpathia	Metasomatic rocks (after various effusives) with quartz, dickite, alunite	Fishkin (I) (1963)
Vilotuĭsk group of fluorite deposits (Eastern Transbaĭkal area)	Veins of chalcendony-like quartz and fluorite (in Lower Paleozoic granites) containing up to 40% nacrite	Polyakova (1965)
Novo-Pavlovsk fluorite deposits in Western Transbaĭkal area	Lens of chalcedony-like quartz (amidst syenites) with fluorite, pyrite, nacrite, hematite	Zvyagin, Scheflov (1962)
Mazad village in Daghestan	Quartz vein (in metamorphic rocks) with pyrite, chalcopyrite, galena, sphalerite; in its drusy cavities – rectorite (allevardite) cinnabar, native silver, cerargyrite	Ivkin *et al.*(1959)
Crimea	Quartz veins (in a phyllite-aleurolitic Tauric flysch) with tosudite (component of alushtite) and rare segregations of carbonate, cinnabar, hematite	Logvinenko, Frank-Kamenetskiĭ (1961)

deposits, igneous rocks and in sedimentary rock masses, which happened to be at considerable depths. In all cases, these minerals could have been deposited from meteoric solutions heated at depth. Depending upon the nature of the latter, different hydrothermal clay minerals were formed.

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Résumé—Des études sur les miné raux argileux en U.R.S.S. effectuées surtout depuis 1960, sont résumées ici. Des techniques employant la diffraction d'électrons ont été spécialement développées et utilisées de façon extensive. Des méthodes ont été développées pour l'enregistrement simultané de courbes DTA et autres propriétés. Les différents types sous lesquels se manifestent les phyllosilicates ont été étudiés intensément par Zvyagin et autres chercheurs. Une grande quantité d'informations nouvelles sur la structure et la morphologie d'halloysites a pu être obtenue par des méthodes optiques et employant les électrons. Les autres minéraux argileux qui ont été étudiés de manière extensive sont les allophanes, les kaolinites, les chlorures ("donbassite" est préféré à "sudoite" pour les variétés dioctraédriques), l'alushtite (ou tosudite), le monothermite et les différentes argiles porteuse de zinc. Une grande attention a été donné aux propriétés physico-chimiques et colloido-chimiques. L'apparition naturelle et l'altération des argiles et des minéraux argileux sont décrites pour les zones climatiques et d'oxidation, pour les sols, et pour les sédiments récents et anciens. Les premiers minéraux argileux d'origine hydrothermique sont décrits.

Kurzreferat—Es wird ein Überblick über die Tonmineralforschung in der Sowjet Union, vor allem seit 1960, gegeben. In erster Linie sind Elektronen-beugungstechniken entwickelt und in weitem Umfang angewendet, worden. Es wurden Methoden für die gleichzeitige Aufzeichnungen von DTA Kurven und anderer Eigenschaften entwickelt. Der Polytypismus von Phyllosilikaten wurde von Zvyagin und anderen gründlich studiert. Durch elektronen-optische Methoden konnten neue Erkenntnisse über den Aufbau und die Morphologie von Halloysiten gewonnen werden. Andere eingehend untersuchte Tonminerale umfassen Allophene, Kaolinite, Chlorite (für oktahedrale Sorten wird "Donbassit" an Stelle von "Sudoit" vorgezogen), Alushtit (oder Tosudit), Monothermit und verschiedene zinkhaltige Tone. Grosse Beachtung wurde den physikalisch-chemischen und kolloid-chemischen Eigenschaften geschenkt. Das natürliche Vorkommen und die Veränderungen des Tons und der Tonminerale in Verwitterungs-und Oxydations-zonen, in Böden und in neueren sowie in alten Ablagerungen werden behandelt. Primäre Tonminerale hydrothermalen Ursprungs werden beschrieben.

Резюме—Приводится сводка работ, предпринятых в СССР по глинистым минералам, главным образом с 1960г. Преимущественно усовершенствовали методы злектронной диффракции и они широко применяются. Усовершенствованы методы одновременной записи кривых DTA

и других характеристик. Звягин и другие интенсивно исследовали политипизм филлосиликатов. Многие новые сведения о структуре и морфологии галлаузитов были получены, пользуясь приемами электронной оптики. Другие глинистые минералы, которые изучались в значительной степени, включают аллофаны, каолиниты, хлориты (причем предпочитают пользоваться донбасситом, а не судоитом для диоктоздрических разновидностей, алуштит (или тосудит), монотермит и разные цинк-содержащие глины. Много внимания уделяется физикохимическим и колоидно-химическим свойствам. Природное нахождение и изменение глни и глинистых минералов описывается в зонах выветривания и окисления, в почвенных слоях, а также в недавних и древних осадках. Описываются первичные глинистые минералы гидротермального происхождения.