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CHLORITE-ILLITE TONSTEIN IN HIGH-RANK COALS FROM QUEENSLAND, AUSTRALIA: NOTES ON REGIONAL EPIGENETIC GRADE AND COAL RANK

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ABSTRACT. The crystals in crystal tonsteins that occur in coals of semi-anthracitic rank in Queensland consist of dioctahedral illite and chlorite, while kaolinite, the usual constituent, is absent. The inference of an epigenetic-authigenic origin of most of the chlorite and illite at the expense of kaolinite is supported by the predominance in the illite of the 1M polymorph. The minor 2M illite, twinned albite, and possibly some of the chlorite are of detrital origin.

The chlorite-dioctahedral 1M illite assemblage in tonsteins and other argillaceous rocks is related to the rank of the associated coals, both being controlled by regional epigenesis or burial metamorphism. The appearance of these minerals in argillaceous rocks associated with coals metamorphosed to or beyond low-volatile bituminous rank is in agreement with previous work; it may be tentatively correlated with at least the quartz-laumontite subfacies of burial metamorphism. Persistence of kaolinite in tonsteins in many high-rank coals is ascribed to compositional factors.

INTRODUCTION

Tonsteins have been described from coals occurring in widely different areas and geological ages. They are mudstones without appreciable directive fabric, essentially consisting of kaolinite, which may occur in a very fine-grained form as matrix, or in pellets (*Graupen*), or as crystals of various sizes.[1] Such crystals, present as columnar and tabular shapes or in vermicular or sheaf-like aggregates, characterize a distinctive type, crystal tonstein. Agreement is lacking on several aspects of the origin of tonstein—in particular, on the origin of the material and on the form in which it was transported and deposited (kaolinite crystallization from aqueous solution or gel, illite-rich mud, or volcanic detritus). Recent summaries of current views on the subject have been given by Moore (1964), Scheere (1958), Stadler (1962), and Eckhardt and Von Gaertner (1962).

Whatever the origin of tonstein, there seems to be substantial agreement that formation of the kaolinite macrocrystals took place under acid conditions in a still-unconsolidated medium, that is, not later than early diagenesis (see, for example, Pietzner, Teichmüller, and Teichmüller, 1962; or, for a dissenting view, Bouroz, 1962). The macrocrystals in practically all described tonsteins consist essentially of kaolinite, sometimes with illite lamellae; the fact that they may be recognized by their characteristic texture makes them a convenient subject for study of mineralogical changes due to epigenesis (late diagenesis).

[1] The "flint clay" partings in Pennsylvanian coals (from the eastern United States) include many tonsteins (see Huddle and Patterson, 1961).

A common effect of regional epigenesis on argillaceous sediments is the tendency for montmorillonite and kaolinite to disappear progressively in shales of increasing age, owing to replacement of authigenic illite and chlorite (Grim, 1953). According to Eckhardt (1958), formation of muscovite-illite from montmorillonite and kaolinite will begin under conditions of pressure and temperature too low for formation of significant chlorite.

In recent years tentative facies of "regional epigenesis and metagenesis", or of "burial metamorphism", have been erected to cover this transition between diagenesis and the greenschist facies of low-grade regional metamorphism.

Kossovskaya and Shutov (1963) distinguish "stages" of progressive mineral alteration in the range of regional epigenesis and lowest-grade regional metamorphism. Approximately covering the same range of burial metamorphorphism, mineral assemblages diagnostic for the zeolite facies and the prehnite-pumpellyite metagraywacke facies are now recognized in rocks of appropriate composition, such as pyroclastic rocks and volcanic graywackes (Coombs, 1961).

Regional epigenesis and coal rank.—Increase in rank—that is, progressive regional metamorphism—of coal, a rock very sensitive to increases in temperature and to a lesser extent pressure (Teichmüller and Teichmüller, 1954) takes place concurrently with epigenetic change in the surrounding sediments. The correlation between the zones of progressive changes in coals and in other sediments is insufficiently established. Logvinenko (1956) relates the progressive late-diagenetic (epigenetic) effects on the mineralogy of argillites and cements of granular rocks in the Donbas and the Dnieper-Donetz basin to the degree of metamorphism of the coal. Three zones are distinguished:

1. Zone of normal epigenesis; region of long-flame, gas, and brown coals.
2. Zone of progressive epigenesis; region of coking coals.
3. Zone of incipient metamorphism; region of lean coals and anthracites.

Alteration of clay minerals to sericite (and chlorite) is predominant in the third zone, which is considered transitional to the greenschist facies. Quinn and Glass (1958) found anthracite and low-rank meta-anthracite of the Narragansett basin, Rhode Island, associated with rocks assigned to the muscovite-chlorite subfacies of the greenschist facies, and a higher-rank meta-anthracite with rocks of the biotite-chlorite subfacies.

These studies provide a preliminary framework of correlation. However, the appearance and nature of authigenic minerals formed during regional epigenesis still is to an appreciable extent determined by the mineralogical composition of the initial rocks; with progressive metamorphism this factor decreases in importance (Kossovskaya and Shutov, 1963).

It is thus desirable to study the progressive epigenetic transformation with reference to coal rank in rock types that not only occur in close association with coal, but in which relevant features of the pre-epigenetic mineralogy are apparent from textural or other evidence. Crystal tonsteins, with their characteristic initially essentially kaolinitic macrocrystals, seem to answer these requirements.

Illite and chlorite in previously described tonsteins.—Minor lamellae of illite are intergrown with the kaolinite macrocrystals of many tonsteins; the name "leverrierite" has been variously used for this illite (Schüller, 1951; Schüller and Grassman, 1954; Hoehne, 1957) and for the kaolinite-illite intergrowths as a whole (for example, Kulbicki and Vetter, 1955). "Leverrierite" is currently understood as merely an aggregation form of illite, with 2M (Saalfeld, 1959) or 1Md (Eckhardt and Von Gaertner, 1962) structure. Grim (1953), Fleischer (1956), and Saalfeld (1959) recommend that the name "leverrierite" be abandoned.

Schüller and Hoehne (1956) regard occasional essentially illite prisms and worms of some tonsteins as syngenetic; most "leverrierite" lamellae they regard as relics of kaolinization of such crystals (Schüller, 1951; Schüller and Hoehne, 1956; Hoehne, 1957). The majority of authors regard the lamellae mainly as relics of detrital illite or mica clays (Kulbicki and Vetter, 1955; Scheere, 1958; Stadler, 1962; Eckhardt and Von Gaertner, 1962).

Bouroz (1962) gives the only description of illite formed during progressive regional metamorphism as a pseudomorphic replacement of kaolinite crystals—in three "meta-tonsteins" in anthracites from central France.

Chlorite has been reported from only a few tonsteins (Schüller, 1951; Hartlieb, 1962; Herbst, Koerner, and Stadler, 1962; Kimpe, 1962; Eckhardt and Von Gaertner, 1962). There is no evidence of a relation between the occurrence and the amount of chlorite and the rank of the associated coals; the latter varies from high-volatile bituminous to anthracitic. Thus there is no indication of an epigenetic origin of the chlorite, which is generally regarded as either detrital or an alteration product of mica.

THE CHLORITE-ILLITE TONSTEINS

The two tonsteins described below were found in coals from the lower portion of the Upper Permian Baralaba Coal Measures of the southeastern part of the Bowen basin, central eastern Queensland. The investigated material comes from cores of a bore drilled by the Geological Survey of Queensland through the coal-measure sequence at Baralaba, some 80 miles southwest of Rockhampton. The sequence contains eleven coal seams of semi-anthracitic rank according to the A.S.T.M. (Am. Soc. Testing Materials) classification. Clean coal composites typically contain 11 to 14 percent volatile matter (dry, ash-free) and 90.5 to 91.5 percent carbon (d.a.f.).

Of these, the tonstein-bearing Dunstan seam is the seventh; clean coal composites of this 14½ foot thick seam and the underlying 3 foot thick sub-Dunstan seam have respectively 11.7 and 12.7 percent volatile matter (d.a.f.) and 91.5 and 91.0 percent carbon (d.a.f.). The Dunstan tonstein occurs at 1067 feet as a band some 5 centimeters thick, in the middle of the coal seam. It is dark colored owing to the presence of about 55 percent by volume of coal material.

Near the base of the coal-measure sequence a coal seam, 2 feet thick, underlies the so-called Tuffmarker horizon. The 8 centimeter thick tonstein occurring in the middle of this coal seam at 1665 feet will be referred to as the "Tuffmarker" tonstein. It is of gray (locally pale brownish gray) color, and grains up to one half millimeter in size are visible in hand specimen; it is rather massive, but contains a few thin, more carbonaceous laminae.

Petrography.—The main mineral constituents of both tonsteins are crystals of brownish pleochroic mica, with pale green chlorite occurring in lamellae up to 15 microns wide or intimately intergrown with the illite (pl. 1). The crystals attain diameters of over 5 millimeters and are columnar, tabular, or vermicular, the basal plates occasionally showing six-sided outlines; generally the crystals are somewhat rounded and diverging. "Sheaf-shaped" ends are common. Chlorite and illite account for virtually all the layer silicate present.

Other constituents in the Tuffmarker tonstein are: albite in subangular grains up to one quarter millimeter in diameter, in part with polysynthetic twinning according to the albite law, and in various stages of sericitization; apatite in columnar crystals of up to 150 microns; very little carbonate. Interstitial carbonaceous material is present in varying but usually very small amounts.

The Dunstan tonstein, according to a volumetric microlithotype analysis of a polished grain mount, contains 54 percent coal (35 percent vitrite, 10 percent intermediates, 9 percent fusite), 6 percent carbargillite, 29 percent layer silicate, and 11 percent carbonate, with traces of pyrite and quartz. The illite-chlorite crystals are locally set in a coaly matrix. Siderite is found in the crystals, possibly as a replacement. Sporadic quartz grains up to one quarter of a millimeter in diameter are present, as well as some apatite; very minor calcite occurs in veins.

Kaolinite was not detected in either sample.

Chemical composition.—A chemical analysis of the Tuffmarker tonstein, made by rapid methods, is as follows: SiO_2, 43.7; TiO_2, 0.97; Al_2O_3, 28.6; Fe_2O_3, 0.51; FeO, 4.94; MnO, tr.; MgO, 1.88; CaO, 1.28; Na_2O, 1.48; K_2O, 4.37; P_2O_5, 1.05; C[2], 1.61; H_2O^+, 7.4; H_2O^-, 2.60; total, 100.4.

[2] Essentially organic carbon; very minor carbonate.

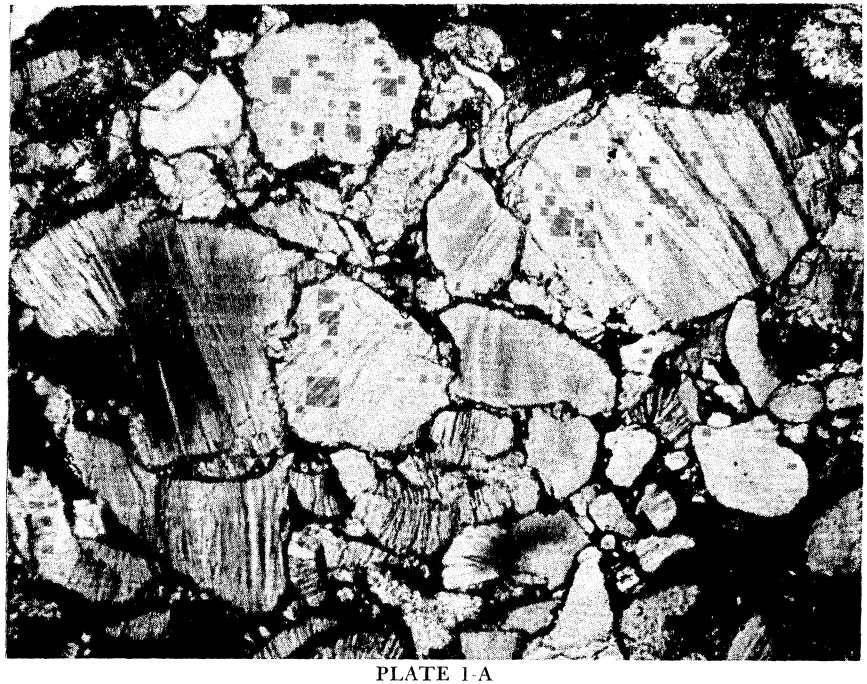

PLATE 1-A

PLATE 1 B

Reflecting the illite- and chlorite-rich mineralogical composition, this chemical composition differs from that of kaolinite crystal tonsteins, notably in its high SiO_2/Al_2O_3 ratio, FeO content, and K_2O content.

The molecular SiO_2/Al_2O_3 ratio, 2 in pure kaolinite, is 2.59; however, if as a simplification all the Na_2O is assumed to be present as albite, and the corresponding amount of Al_2O_3 and SiO_2 are subtracted from the totals, the ratio for the rest of the rock becomes 2.27. These ratios are similar to many "leverrierite" (illite)-rich tonsteins (Hoehne, 1957; Scheere, 1958); the average ratio for 74 tonstein samples of all types from the Saar region is 2.59 (Schüller and Hoehne, 1956).

The ferrous iron content is comparatively high; in analyzed illites it is less than 2 percent and practically always appreciably lower than the magnesium content. Consequently, the atomic ratio $(Fe^{+2} + Fe^{+3}) / (Fe^{+2} + Fe^{+3} + Mg)$ in the chlorite at least equals that of the whole rock, 0.6. Some of the iron may originally have been present as siderite or may have been released during epigenesis. The calcium and phosphorus contents are almost exactly in the same proportion as in apatite, indicating that the other constituents are virtually calcium-free.

Mineralogy of the chlorite and illite.—The chlorite and illite were studied optically, by X-ray powder diffraction and by differential thermal analysis. Samples heated to 550°C (2 hours) and other samples glycolated and extracted with hot 2N HCl (2 hours) were also investigated. A mineral concentrate was used for the Dunstan tonstein.

The *chlorite* is pale green, optically negative, with comparatively strong birefringence, and refractive indices appreciably higher than those of the mica (the occurrence as thin lamellae precludes accurate measurement). The chlorite is unequivocally identified by the changes in the X-ray diffraction pattern on heating and acid treatment: the (001) reflection is much enhanced on heating to 550°C, while the other basal reflections decrease or disappear; glycolation has no effect on the spacings; the whole pattern disappears on treatment with hot 2N HCl, which also confirms the absence of kaolinite. The (001) and (003) reflections of the unheated chlorite are weak, and the (002) and (004) reflections strong: $F_{(001)}/F_{(002)} \cong 0.2$, and $F_{(003)}/F_{(004)} \cong 0.4_5$ for both tonsteins, indicating high iron content (Brindley, 1961) in agreement with the chemical data. The value of $d_{(001)}$ (calculated from $d_{(002)}$, $d_{(003)}$, and $d_{(004)}$) is 14.0 to 14.1 Å. According to data of Shirozu (in Brindley, 1961, p. 270) this indicates 3 to 4 $[Al]^4$ ions per formula unit, that is, an aluminous chlorite.

PLATE 1

A. Chlorite lamellae in vermicular crystals of pleochroic (brownish in vertical position) illite in the Dunstan tonstein; the interstitial material is mainly coal. Plane-polarized light, 330 x.

B. Same as A, but with crossed nicols.

The *illites* are brownish pleochroic. They have a maximum birefringence of approximately 0.030, and refractive indices $n_\gamma = 1.590 \pm 0.002$ and $n_\alpha = 1.559 \pm 0.002$. The structure is dioctahedral, as indicated by the spacing of (060) at 1.495 to 1.505 Å. The X-ray powder diffraction patterns of the illites from the two tonsteins are rather different as to the basal spacings; these are given in table 1 for the untreated or the hot 2N HCl-treated samples.

The (001) and (003) reflections of the Tuffmarker illite have "tails", which are much less apparent in the Dunstan tonstein, toward the low-angle and the high-angle side respectively. Glycolation of the former reduces the intensity of the (001) reflection and displaces it to 9.9 to 10.0 Å, while (003) is somewhat sharpened and displaced to 3.36 Å; this suggests the presence of a minor amount of mixed layer clay with less than 20 percent expandable layers (Weaver, 1956). On heating to 550°C the (001) reflection loses its low-angle tail and decreases to 10.2 Å, while (002) and (003) are enhanced, the latter increasing to 3.36 Å. The fact that (001) does not shift to 10 Å on heating —as would be expected if all the interstratified layers were expandable— may indicate that a minor amount of 14 Å chlorite, on the average less than 10 percent, is randomly interstratified with it. The same may be true of the illite of the Dunstan tonstein, the diffraction pattern of which is hardly affected by heating to 550°C, apart from some enhancement of the (003) and (005) reflections relative to (001). Glycolation does not change the pattern: this illite contains no or negligibly few expandable layers.

Reflections at 3.07 Å and probably also at 3.68 Å — the latter obscured by an albite reflection—indicate that the illite in the Tuffmarker tonstein is essentially the 1M (not 1Md) polymorph; a very minor amount of 2M is present (Yoder and Eugster, 1955). In the Dunstan tonstein the predominant polymorph is also 1M (peaks at 3.10-3.11 and 3.68-3.69 Å (with subordinate 2M.

Differential thermal analysis of the Tuffmarker tonstein gave endothermic reactions at 120, 155 to 160, 575 to 580 (strong) , and 750 to 755°C (heating to 960°C) . An acid-treated (that is, chlorite-free), oven-dried

TABLE 1

Basal spacings for illite of Tuffmarker and Dunstan tonsteins

(001)	Tuffmarker peak (d in Å)	I	Dunstan peak (d in Å)	I
(001)	10.3–10.4	s	10.3–10.4	s
(002)	5.00–5.06*	w	5.14–5.16	w
(003)	3.34	s	3.41–3.42	m
(005)	2.016–2.023*	m	2.054–2.060	w

* Possible double peaks.

sample has a strong endothermic peak at 560°C, and others at 740 to 745, 850, and (?) 960°C; with exothermic peaks at 895 to 900 and (?) 980 to 985°C. The absence of the peaks above 800°C from the untreated sample could be due to masking effects of chlorite. A similarly treated sample of the Dunstan tonstein has a strong endothermic peak at 535 to 540°C and one at 980°C, and a marked exothermic peak at 995 to 1000°C.

Endothermic peaks at 110 to 150, 550 to 600, and 850 to 900°C, and an exothermic peak at 900 to 1000°C are characteristic of illite, particularly the 550 to 600°C peak, but some fine-grained muscovites have endothermic peaks at 700 to 750°C (see Bradley and Grim, 1961, fig. V; and Grim, 1953, p. 235). This suggests that the dioctahedral mica consists of illite, in the Tuffmarker tonstein possibly in association with minor amounts of muscovite.

DISCUSSION

The major points of petrological interest which have emerged are the presence of chlorite as a major constituent of the crystal tonsteins together with a dioctahedral illite, and the absence of kaolinite—the characteristic constituent of the macrocrystals in crystal tonsteins.

The dioctahedral illite has been shown to be mainly of the 1M polymorph, with minor 2M material. Yoder and Eugster (1955) found in their experimental studies of stability relations of muscovite polymorphs that the 1Md polymorph is a metastable form that is subsequently ordered, with increasing temperature and duration of heating, into the 1M structure, which is stable below the temperature range 200 to 350°C. However, according to Weaver (1959), 1Md material derived by degradation of 2M dioctahedral mica will easily be reconstituted to the 2M structure by replacement of K lost during weathering: 2M structure can thus be regarded as indicating a detrital origin in illites in sedimentary rocks. Assuming that the stability ranges found experimentally for the muscovite polymorphs by Yoder and Eugster apply to illite (Velde and Hower, 1963), the 2M polymorph will not form from the 1M polymorph during progressive metamorphism below the temperature range 200 to 350°C. Temperatures below 200°C are held by Huck and Karweil (1955) to suffice in general for anthracite formation: thus the minor amounts of 2M illite in the tonsteins must represent detrital, or degraded and reconstituted dioctahedral mica.

Conversely, 1M illite would not be expected to form from detrital and degraded dioctahedral mica. Its presence in the tonsteins is taken to indicate authigenic origin, that is, formation during epigenesis (late diagenesis), either by progressive ordering of syngenetic 1Md illite or by replacement of kaolinite.

The presence in the tonstein of constituents of detrital origin such as angular grains of polysynthetically twinned albite, as well as 2M illite, suggests that some of the chlorite present may be of detrital origin

as well. However, the mode of occurrence of the chlorite as a major constituent of the macrocrystals, in intimate intergrowth with illite, cannot be accounted for by a detrital or early diagenetic origin: the major part of the chlorite must be considered authigenic-replacive, formed during epigenesis (late diagenesis) at the expense of the original constituent of the macrocrystals, that is, kaolinite (see also Bouroz, 1962).[3]

The presence of the epigenetic assemblage chlorite-dioctahedral illite in mudstone associated with coal of semi-anthracitic rank is in accord with correlations between epigenetic grade and coal rank cited earlier. Logvinenko (1956) gives the following assemblages in argillites and cements of sandstones in the "zone of incipient metamorphism" which is correlated with development of lean coals and anthracites: sericite, sericite-quartz, sericite-quartz-carbonate, carbonate, and sericite-quartz-chlorite. Quinn and Glass (1958) note the association of anthracites having 5 to 8 percent volatile matter (d.a.f.) with a fine-grained muscovite-paragonite-chlorite assemblage, regarded as metamorphosed in the muscovite-chlorite subfacies of the greenschist facies. Kossovskaya, Shutov, and Alexandrova (1964) ascribe the wide distribution of chlorite-dioctahedral hydromica argillites and the absence of kaolinite even from the floors of coal seams (which elsewhere are kaolinite-rich) in the lower part of the Vorkuta Series (Petchora basin) to reworking in deep epigenesis and metagenesis; coal ranks in this series range up to semi-anthracitic. Eckhardt (1958) regards sericite and possibly chlorite in a marine shale associated with medium-volatile bituminous coals in the Ruhr area, as formed from kaolinite and montmorillonite. Epigenetic illite in pseudomorphs after kaolinite crystals is reported from metamorphosed tonsteins in some anthracites from central France (Bouroz, 1962).

Eckhardt and Von Gaertner (1962), on the other hand, reported no authigenic chlorite in their mineralogical study of a number of tonsteins from Western Germany associated with coals of ranks up to semi-anthracite. These authors did, however, establish a relation between degree of order in the kaolinite of the tonsteins and their stratigraphic position and coal rank: tonsteins of the Westphalian A and B contained well-crystallized kaolinite, those from the Westphalian C and the Wealden coals more disordered kaolinite tending toward "fireclay-

[3] Since this paper was written, a kaolinite tonstein was found some 30 miles southeast of Baralaba in the medium-volatile bituminous (29-30 percent volatile matter, about 86.5 percent C, d.a.f.) "C" seam near Moura, which has been tentatively correlated with the Dunstan seam at Baralaba. The newly discovered occurrence is a pellet (Graupen) tonstein grading to pellet-crystal tonstein, after Schüller and Hoehne (1956) and Eckhardt and Von Gaertner (1962), and up to 5 centimeters thick. Rounded pellets of generally very fine-grained kaolinite, only locally up to 100 microns, and occasional larger crystals are embedded in a coal matrix. Apart from very sparse large illite crystals, the clay mineral is kaolinite, indicating that the epigenetic alteration of kaolinite into chlorite and illite is negligible at this degree of coalification.

mineral". They considered the degree of order to reflect differences in temperature of coalification.

The absence of authigenic chlorite and dioctahedral illite and the persistence of kaolinite in many tonsteins subjected to deep epigenesis, that is, from coals of low-volatile bituminous rank and over, particularly when associated shales are chlorite-rich, must be due to insufficient concentration of required chemical components, notably potassium and iron. Alteration of kaolinite into dioctahedral illite during epigenesis will take place only with potassium, while its chloritization requires iron or magnesium (Grim, 1953; Eckhardt, 1958; Kossovskaya, Shutov, and Drits, 1963; Garrels and Christ, 1965). Kossovskaya and Shutov (1963) have proposed a correlation between critical minerals or mineral assemblages—termed "facies of regional epigenesis and metagenesis"—formed in five compositional "families" of terrigenous rocks within each of four successive "stages": "initial epigenesis", "deep epigenesis", "early metagenesis", and "late metagenesis". In the stage of deep epigenesis the characteristic "facies" are: quartz-dickite (family of quartz-kaolinite rocks), chlorite-hydromica (family of polymictic lithoclastics), quartz-albite-hydromica (family of "acid arkoses"),[4] and laumontite "facies" (families of "intermediate arkoses"[5] and of volcanic graywackes). According to this correlation the mineral assemblages of the chlorite-illite tonsteins could be formed under conditions at least of the quartz-laumontite subfacies of Coombs' (1961) zeolite facies.

When insufficient K and Fe or Mg are available dickite might thus be expected to form at a similar epigenetic or burial metamorphic grade in some pure kaolinite tonsteins (Kossovskaya and Shutov, 1963; Kossovskaya, Shutov, and Drits, 1963). The studies of Eckhardt and Von Gaertner seem to indicate that kaolinite may persist, but it would be advisable to be on the lookout for dickite—which may easily be overlooked because of the similarity of its X-ray diffraction pattern to that of kaolinite—in tonsteins from high-rank coals. Nevertheless, it is to be expected that some other tonsteins from semi-anthracites and anthracites will, on closer examination, turn out to contain authigenic chlorite and dioctahedral 1M illite.

It may be concluded that the replacement of kaolinite by chlorite and dioctahedral 1M illite does not of necessity take place once a certain grade of regional epigenesis, as indicated by coal rank, is attained. However, when the compositional requirements are satisfied, the transformation of kaolinite into the chlorite-dioctahedral 1M illite assemblage in crystal tonsteins and other initially kaolinite-rich rocks is believed to represent an important stage in their epigenetic transformation. Further attempts to establish a correlation with the rank of the associated coals should contribute toward the clarification of the relation between coal

[4] With plagioclase up to An_{20} and/or potassium-rich feldspars.
[5] With plagioclase up to An_{15}.

rank and facies of regional epigenesis and low-grade regional metamorphism.

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