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Colour zones and the transition from diagenesis to low-grade metamorphism of the Gedinnian shales around the Stavelot Massif (Ardennes, Belgium)

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Abstract

Red to bluish gray colour transformations of the Gedinnian shales around the Stavelot Massif are shown to result from progressive metamorphism in the fields of diagenesis to the lower epizone.

Muscovite compositions indicate conditions of low pressure metamorphism and reveal the existence of a temperature gradient of 115° between the northern and southern border of the Massif.

Illite crystallinity data, as well as the colour zones of the Upper Salmian rocks indicate that the metamorphism is related to the Hercynian orogeny.

Introduction

Low-grade metamorphic rocks of the Upper Salmian (Tremadoc) along the southern border of the Stavelot-Venn Massif are characterized by associations of chloritoid, spessartine and sometimes andalusite. They have been described in detail by THEUNISSEN (1970), KRAMM (1973, 1976) and FRANSOLET et al. (1977). It is unclear wether this metamorphism is related to the Caledonian or the Hercynian orogeny.

Spessartine garnets, occurring in the Gedinnian conglomerates just South of the Massif, at least prove the existence of an Hercynian metamorphic period (SCHREYER, 1975). Both the Upper Salmian and the conglomerates of the Gedinnian, however, are less suitable for a regional investigation of the metamorphism in the area since only the more manganiferous portions have developed metamorphic assemblages. The manganiferous rocks of the Upper Salmian occur only in the cores of synclinal units, while in the conglomerates of the

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Gedinnian only conglomerates with pebbles derived from the Upper Salmian show the effects of the metamorphism.

The Gedinnian "Oignies shales" are of special interest since they can be traced nearly continuously around the Massif. Recognizable metamorphic assemblages are absent but the shales show a systematic change in colour from deep red along the northern border of the Massif to bluish gray along the southern border.

The object of this paper is to define metamorphic zones in terms of clay mineral assemblages, to relate these to the observed colour changes and to relate these features in time and space with the Caledonian formations.

Colour zones

In the Gedinnian shales around the Stavelot Massif, three distinct zones could be distinguished on the basis of colours. Whereas red colours occur North and West of the Massif (zone A), the shales are bluish gray in the South (zone C). These zones are separated both on the eastern and western side by a relatively narrow zone (B) of reddish gray shales (see figure 1). It is also remarkable to notice the similarities with colours of the Upper Salmian shales. Not only do the same colour changes occur in these rocks, but where their outcrops are localized near the border of the Massif, we could establish a very good agreement with the colour zones of the Gedinnian rocks. A spectral reflectance curve typical of each zone is given in figure 2.

The results of our petrographical and chemical investigations bearing on the colour of the Gedinnian shales are presented in table 1. No effect of the molar oxidation ratio ($2Fe_2O_3x100/[2Fe_2O_3+FeO]$), the amount of total iron, or the Ti and Mn contents on the colour of the shales could be established from the 33 samplas that were analysed.

Frey (1969 a) reports analogous red to violet colour transformations in a progressive metamorphic sequence of the Glarus Alps. He related these to the incorporation of Ti in the hematite structure. The ilmenite content of our hematite was determined by diffractometer using Co$K\alpha$ radiation in the same manner as Frey (1969 a, p. 103). The data were computed according to Lindsley (1963). The variations are not significant and the large discrepancies between the data based on the (024) and (116) reflections of hematite lead us to doubt the reliability of the method for low Ti concentrations.

The most striking differences are recorded for the grain size of hematite. Whereas homogeneously distributed dusty hematite predominates in the red shales, it becomes increasingly coarser and individualized as hues change to bluish gray. The percentage of "easily reduced» iron, determined by the method of Deb (1950) as modified by Jackson (1958), provides an estimate of the

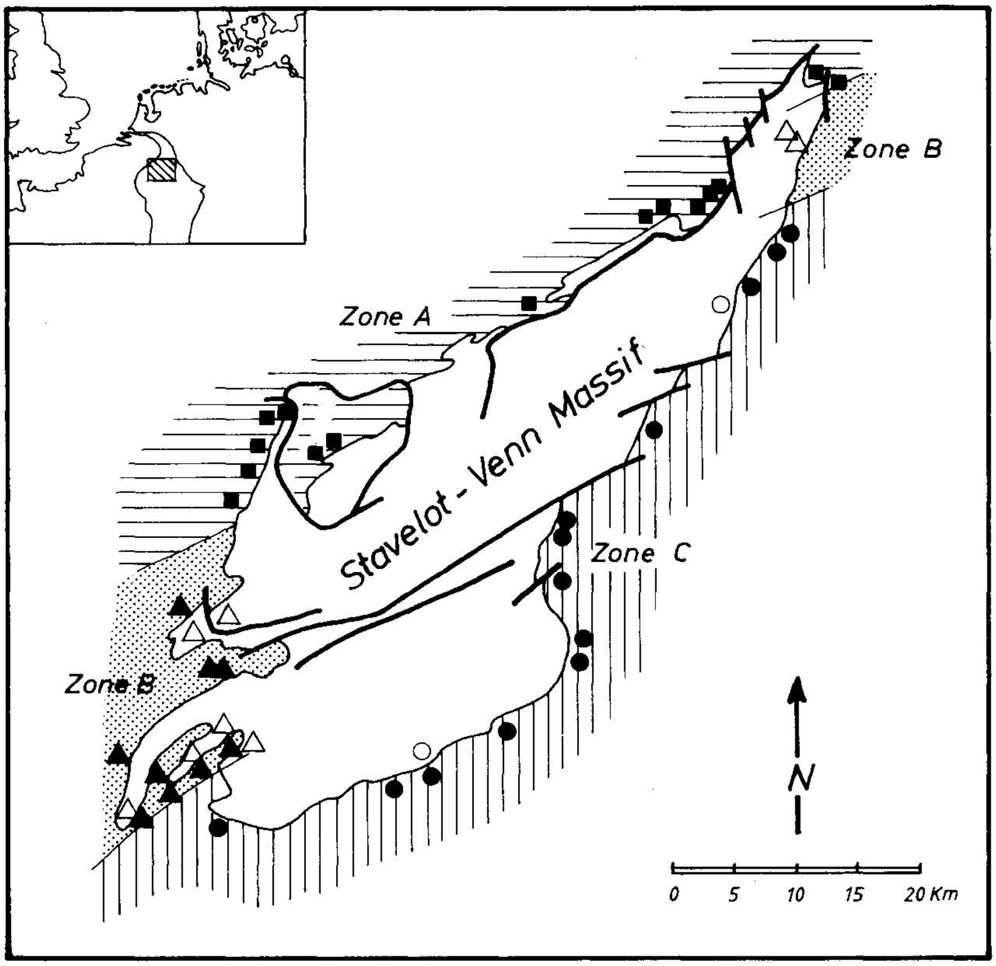

Figure 1 Geological map of the Stavelot-Venn Massif (after F. Geukens). The various hatschings indicate colour zones of the Gedinnian shales.
Sampling localities are indicated by closed symbols for the Gedinnian, open symbols for the Upper Salmian.

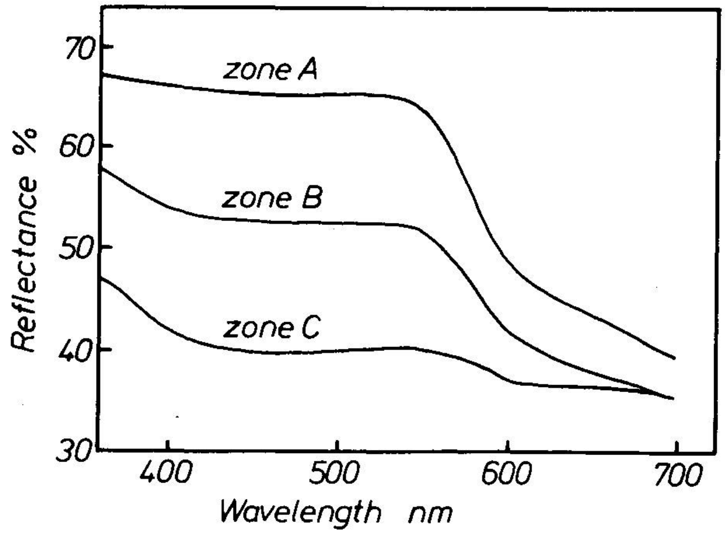

Figure 2 Spectral reflectance curves of powdered samples of the various colour zones.

Table 1 Results of various determinations performed on the Gedinnian shales.

	Zone A [*]	Zone B	Zone C
Rock colour	Dark reddish brown Dark red	Dark reddish gray Grayish red	Dark bluish gray
Index of Rock-colour Chart	2.5 YR 3/2 - 3/4 10 R 3/3 - 3/4	7.5 R 4/1 7.5 R 4/2	5 PB 4/1
Weight % total iron as Fe_2O_3	4.52 - 8.42 6.80 ; (7)	4.83 - 7.69 6.48 ; (13)	5.09 - 8.74 7.30 ; (13)
$2Fe_2O_3x100/(2Fe_2O_3+FeO)$ [**]	73 - 96 84 ; (7)	47 - 97 84 ; (13)	45 - 93 73 ; (13)
Weight % TiO_2	0.15 - 0.23 0.20 ; (4)	0.10 - 0.36 0.19 ; (10)	0.13 - 0.35 0.20 ; (7)
Weight % MnO	0.002 - 0.033 0.021 ; (7)	0.003 - 0.029 0.013 ; (13)	0.006 - 0.057 0.020 ; (13)
Hematite $2\theta_{(024)}$ Fe-$K\alpha_1$	69.575	69.617	69.596
Mol per cent ilmenite	9.9 ; (3)	6.8 ; (5)	8.6 ; (5)
Hematite $2\theta_{(116)}$ Fe-$K\alpha_1$	63.461	63.438	63.441
Mol per cent ilmenite	1.8 ; (3)	2.1 ; (5)	2.1 ; (5)
Grain size of hematite	< 1µ	1 - 10µ	4 - 40µ
Weight % "easily reduceable iron" as Fe_2O_3	1.39 - 2.09 1.64 ; (8)		0.33 - 1.33 0.79 ; (8)
Illite crystallinity	8.5 - 13	4 - 5.5	3 - 4
Muscovite b_o	8.991 - 9.000 8.998 ; (8)	8.991 - 9.000 8.995 ; (8)	8.992 - 9.000 8.996 ; (12)
RM average	0.020	0.014	0.021
d(002)	9.987 - 9.995 9.991 ; (8)	9.982 - 9.989 9.986 ; (8)	9.978 - 9.965 9.973 ; (12)
Na/Na+K average	0.071	0.096	0.127

[*] Only samples from the northern border of the Stavelot Massif.

[**] Fe_2O_3 and FEO expressed as molar contents.

Data linked by a dash indicate the range, the other data indicate the average value and the (number of samples).

amount of leachable hematite in the samples. The data indicate a decrease from zone A to zone C. Although these results are largely influenced by the grain size of hematite, and thus confirm the petrographic observations, it is possible that they also indicate a decrease of the hematite content. This is also corroborated by diffractograms which show slightly decreasing peak-areas of hematite.

Mineral parageneses

Mineral parageneses of the three zones were determined by X-ray diffraction with the following results:

Zone A: illite; chlorite; mixed-layer paragonite-illite; traces of albite.

Zone B: muscovite; chlorite; mixed-layer paragonite-muscovite; pyrophyllite in three samples out of nine; the presence of paragonite in two samples was inferred from the occurrence of a reflection at 1.92 Å (00.10).

Zone C: muscovite; chlorite; paragonite; mixed-layer paragonite-muscovite.

All the rocks contain quartz. The presence of the mixed-layer paragonite-muscovite was deduced from the basal reflections at 4.90 Å, 3.25 Å and sometimes 1.96 Å (Frey, 1969b). The reflection at 49–55 Å was not detected. The petrographic study of the shales did not reveal the presence of K-feldspar.

Rock chemistry

Chemical analyses of rocks from the different zones were performed by the method of Voinovitch (1962).

The results are presented in table 2. As can be seen from the SiO_2 contents and the low amounts of CaO, the rocks are quartziferous shales. MnO is practically absent. The high Al_2O_3 content is probably their most striking feature; in an AKNa projection all analyses plot closely together above the muscovite-paragonite tie line.

Rocks with sufficient Al_2O_3 to plot above the garnet-chlorite join on an AFM projection are likely to contain paragonite (Guidotti, 1968). An AFM projection through muscovite of our analyses (figure 3) shows that this is the case. After correction of the analyses of zone C for paragonite, the representative points still plot above the chlorite field. Baltatzis and Wood (1977) have also drawn attention to the importance of the alkali ratio of the rocks as a constraint on the appearance of paragonite in rocks of the same metamorphic grade. Our data on the paragonite bearing samples (analyses 4 to 7), confirm this view to a certain extent, the larger the Na/Na+K the larger is the proportion of paragonite to muscovite.

Table 2 Chemical composition of the Gedinnian shales.

Sample	1	2	3	4	5	6	7
zone	A	A	A	B	C	C	C
SiO_2	66.73	58.27	58.34	66.67	62.61	68.63	61.93
TiO_2	0.89	0.88	0.93	0.80	0.94	0.88	0.94
Al_2O_3	16.17	19.61	18.91	17.23	19.49	17.11	19.36
Fe_2O_3	5.07	7.40	6.01	6.52	6.20	4.85	6.33
FeO	1.34	1.25	1.76	0.36	0.99	0.90	1.19
MnO	0.02	0.03	0.03	0.01	0.02	0.01	0.02
MgO	1.51	1.81	3.66	0.55	1.42	0.61	1.58
CaO	0.10	0.16	0.09	0.10	0.05	0.03	0.05
Na_2O	0.21	0.52	0.34	0.52	0.75	1.08	1.26
K_2O	3.50	3.58	3.52	3.84	4.10	3.16	3.35
H_2O^+	3.91	4.88	5.19	2.63	2.92	2.40	3.01
H_2O^-	0.85	0.99	0.82	0.26	0.16	0.05	0.12
	100.30	99.33	99.59	99.53	99.65	99.71	99.13
Na/Na+K	0.08	0.18	0.13	0.17	0.22	0.34	0.36

Analyst : J. Paenhuys.

Crystallographic parameters of micas and chlorites

Due to the very fine grained nature of the shales we had to resort to X-ray diffraction methods in order to determine the compositional characteristics of the various phases.

Muscovite

Crystallinity and intensity ratio I(004)/I(002)

Illite crystallinity data were obtained by measuring the peak-width of the 10 Å reflection at half height above the background (KÜBLER, 1968; DUNOYER DE SEGONZAC, 1969). Measurements were performed on the fractions ‹2 μ and 2–6 μ. The values, expressed in millimetres, however, depend on the running conditions: CoKα radiation, 1° pro min., paper 1200 mm/h, 2000 cps, TC 1, slits 1°–0.1 mm–1°.

Illite crystallinity measurements have received considerable interest in the recent past as they provide an indication of metamorphic grade in the field of

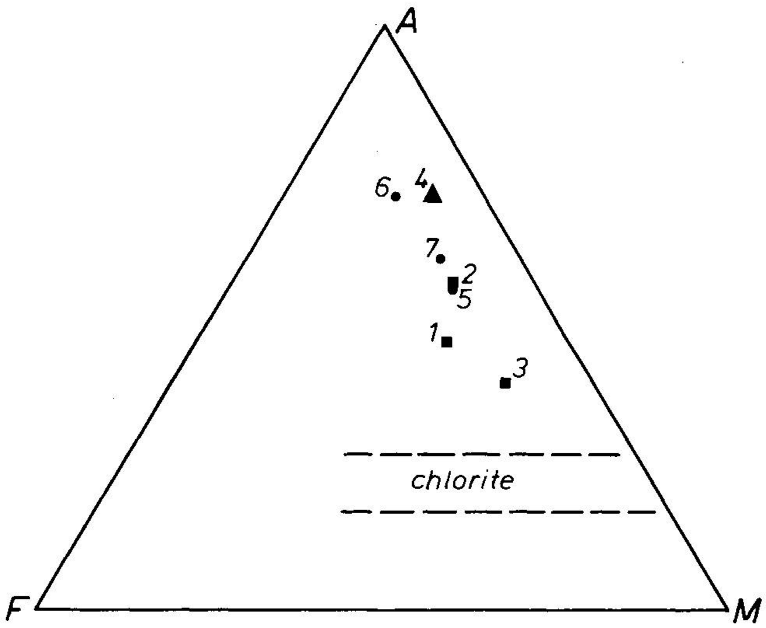

Figure 3 AFM projection of the analyses of the Gedinnian shales.

diagenesis and the lower epizone (DUNOYER DE SEGONZAC, 1970). To afford a comparison of data, Dunoyer de Segonzac kindly provided five illite crystallinity standards. On his scale, the diagenetic/anchimetamorphic boundary and the anchimetamorphic/epimetamorphic boundary correspond to a crystallinity index of 3.5 and 5.5 respectively (5.2 and 8.5 on our scale).

Figure 4 represents our measurements together with the crystallinity scale of Dunoyer de Segonzac.

The results on the fractions <2 μ cover the range of diagenesis to epizone and reflect a progressive increase of illite crystallinity from the north side of the Massif towards the south: in zone A the samples from the northern border have diagenetic characteristics, those of the west side plot in the anchizone field. In zone B the crystallinities have lower epizone values and are transitional to those of the southern margin where the bluish shales possess the highest epizone crystallinities. The transition of diagenesis to anchizone had no influence on the reddish brown colour of the rocks of Zone A.

The transition to the epizone on the contrary coincides with the change to reddish gray (Zone B). The change to bluish gray (Zone C) provides an additional boundary, which in our case coincides with the systematic appearance of paragonite once a crystallinity index of 4 has been attained. The fractions 2–6 μ systematically show better crystallinities, as could be expected from their coarser grain sizes. The results, however, confirm the relation between colour zones and illite crystallinities. The crystallinity data are plotted against the intensity ratio of the peaks at 5 Å and 10 Å, which, according to Esquevin (1969), is pro-

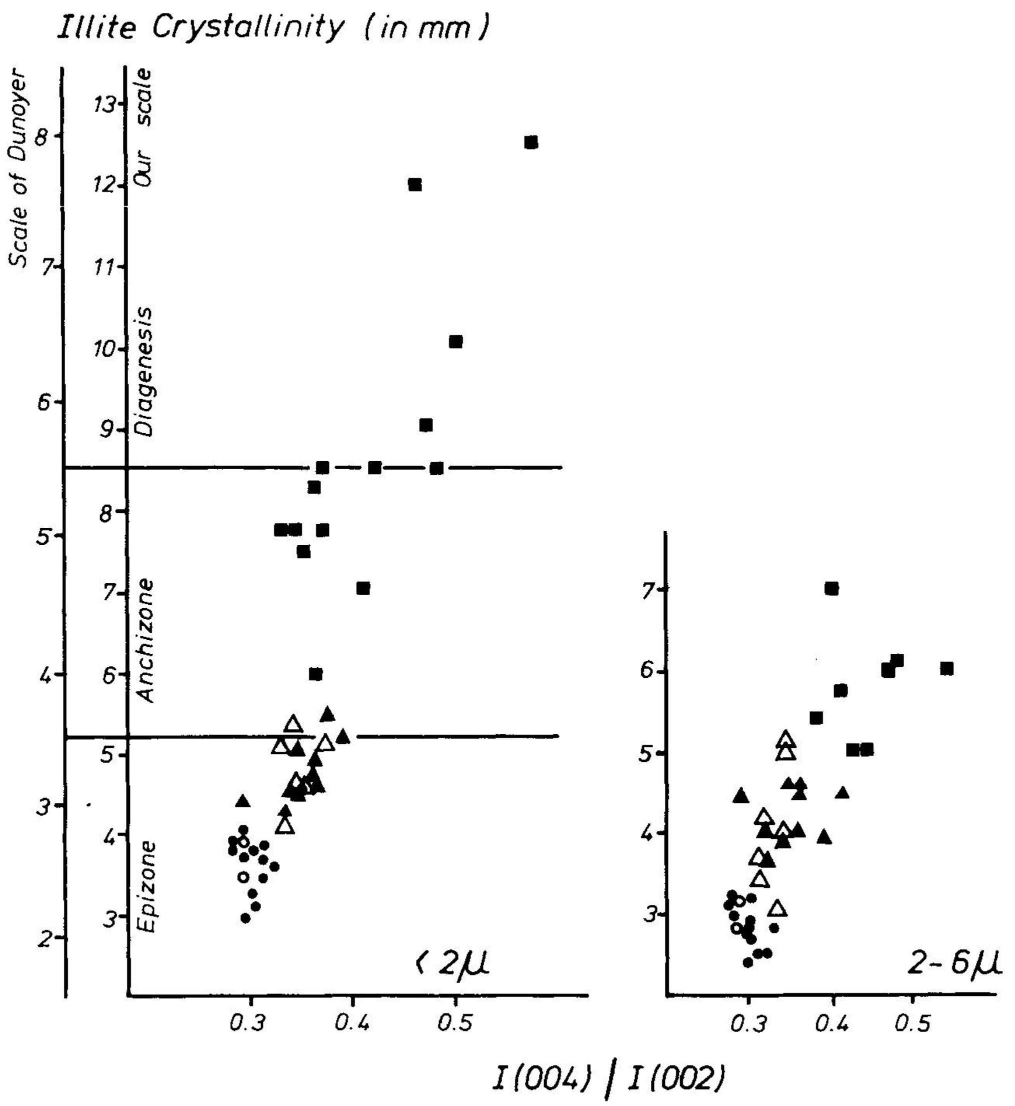

Figure 4 Illite crystallinities of the Gedinnian and Upper Salmian. Symbols represent colour zones and refer to figure 1.

portional to the ratio Al/(Fe+Mg) in the octahedral sites. The results turned out to be relatively constant.

Figure 4 also shows crystallinity data of rocks of the Upper Salmian, sampled near the border of the Massif. The similarity of these values with those of the nearby Gedinnian shows that most, if not all, of the colour and crystallinity changes can be ascribed to a post-Gedinnian event.

Polymorphs

According to Velde and Hower (1963) and Maxwell and Hower (1967) the ratio of polymorphs, 2M/2M+1Md increases with the degree of evolution of the sediments towards metamorphism. In all the rocks studied here the 2M

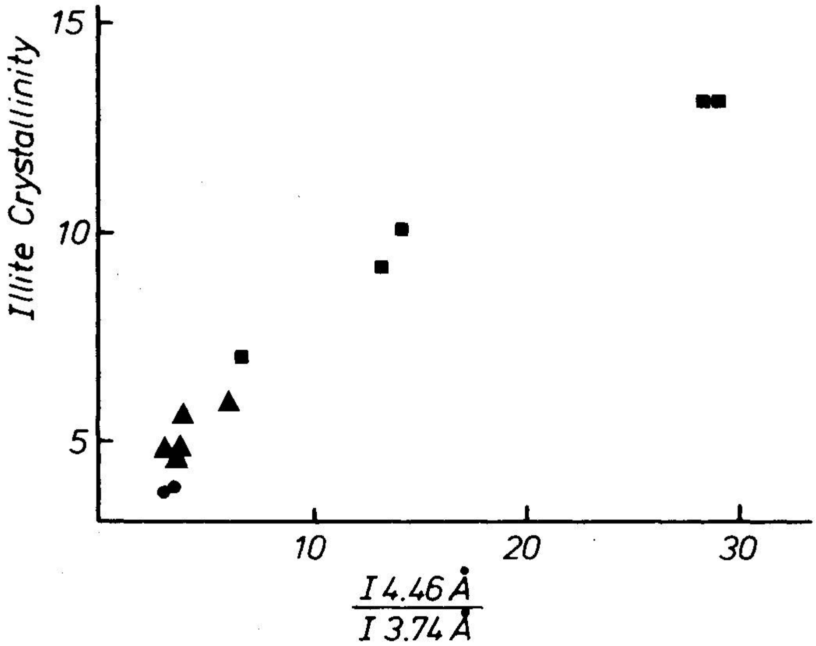

Figure 5 Illite crystallinity versus Intensity ratio of 4.46 Å and 3.74 Å reflections. Symbols refer to figure 1.

structural polytype clearly dominates. An estimation of the 1Md proportion was obtained from the intensity ratio of the reflections at 4.46 Å (2M) and 3.74 Å (2M+1Md) (DUNOYER DE SEGONZAC, 1969). This ratio is plotted against the crystallinity values of figure 5: it shows a decrease of the 1Md proportion as the crystallinity improves, up to epizone at crystallinity 5, where the 1Md proportion remains constant.

Muscovite-paragonite relation

Measurements of the basal spacings of muscovite and paragonite pairs from Zone C rocks were performed by repeated runs across the 00.10 peaks of the micas at goniometer and paper speeds of ¼° pro min. and 600 mm/h, using quartz as an internal standard.

ZEN and ALBEE (1964) have shown that the basal spacings of coexistent mica and paragonite fit the regression equation:

$d(002)_{2M}par. = 12.250 - 0.2634d(002)_{2M}musc. \pm 0.0006$ in Å.

The results of our measurements are shown in figure 6 along with the regression line of Zen and Albee. All the data plot closely together, within the uncertainty limits of the regression line. According to Zen and Albee, such a feature indicates chemical equilibrium between the two micas, provided that the "celadonite" content (i.e. Fe+Mg substitution for Al) of the muscovite is low.

ZEN and ALBEE (1964) also stated that the spacings of mica pairs reflect the metamorphic grade. Our data are comparable with their results for chlorite zone rocks.

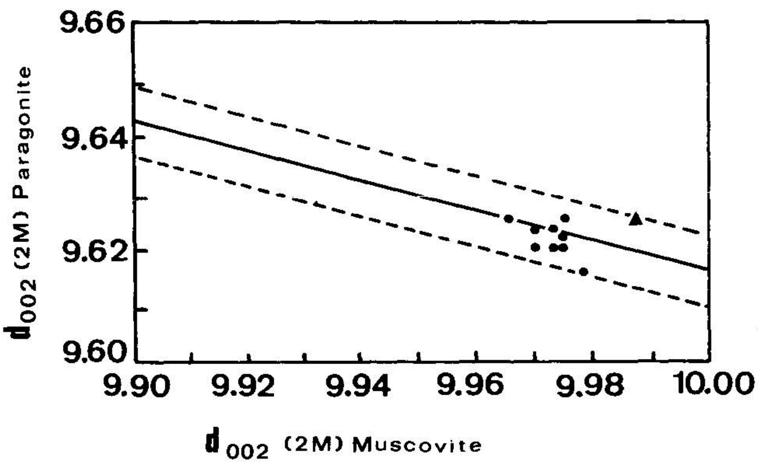

Figure 6 $(002)_{2M}$ spacings of coexistent muscovites and paragonites. Symbols refer to figure 1. The straight line corresponds to the regression line $d(002_{2M}$ par. = 12.250–0.2634 $d(002)_{2M}$ musc. (after ZEN and ALBEE, 1964).

"Celadonite" and "paragonite" contents of muscovites

It is well known that the lattice constant b_o d(010) of muscovite is related to its "celadonite" content, while the basal spacing d(002) is controlled by the alkali ratio (Na/Na+K). CIPRIANI et al. (1968) suggested to use a b_o value of 9.025 Å as a boundary between phengites (b_o<9.025 Å) and muscovites (b_o>9.025 Å). All the muscovites studied here have b_o values between 8.99 and 9.00 and are thus very aluminous muscovites. This scheme is, however, complicated due to mutual interference of the ionic substitutions on the crystallographic constants. The possibilities of X-ray diffraction as a tool for estimation of the composition of muscovite, have been reviewed by GUIDOTTI and SASSI (1976). These authors suggest computing the femic content (RM) and the alkali ratio (Na/Na+K) from the following mathematical relations given by CIPRIANI et al. (1968):

$d_{(002)}$ = 10.023 - 0.316 Na/Na+K) - 0.484 RM RM<0.10

b_o = 8.995 - 0.039 Na/(Na+K) + 0.321 RM

Our measurements together with the compositional data based on these relations are given in table 1. The RM values show up to be fairly constant and prove the low amount of "celadonite" substitution in the muscovites. The mole proportion of paragonite, however, increases from about 7 percent in Zone A to 10 percent in Zone B and approximates 13 percent in Zone C. The largest amounts of "paragonite" substitution in Zone C were observed for those samples containing the smallest modal proportion of paragonite in the mineral assemblage.

GUIDOTTI and SASSI (1976) also reviewed and discussed the influence of pressure and temperature on the muscovite composition. Considering their findings and regarding the barometric scale of SASSI and SCOLARI (1974), the following conclusions can be drawn:

a) the low and roughly constant b_0 values point to conditions which are comparable to "low pressure metamorphism with chlorite zone" of the Hercynian metamorphism in the Eastern Alps (SASSI and SCOLARI, 1974).
b) at low pressure the temperature has no noticeable influence on the "celadonite" content (GUIDOTTI and SASSI, 1976), so that the alkali ratio closely reflects the temperature variations. Since our muscovites and paragonites are close to the binary $KAl_3Si_8O_{10}(OH)_2$ - $NaAl_3Si_8O_{10}(OH)_2$ solid solution, it should be possible to estimate the temperature of crystallization from the solvus of EUGSTER et al. (1972) determined at 2 kb: this yields average values of 300° for Zone A along the northern border of the Massif, 370° for Zone B and 415° for Zone C, indicating the existence of a temperature gradient of 115° from north to south.

Chlorite

The composition of the chlorites was determined by diffractometer according to the method of WETZEL (1973). Al_{tot} was calculated from d(005). The total iron content was determined using the uncorrected Petruk coefficient (I[003]/I[002]+I[004]). Although within each colour zone the results turned out to be fairly consistent, they have to be considered as an approximation. The method does not allow an evaluation of the proportion of Al in tetrahedral and in octahedral lattice positions. The results are:

- Zone A: $(Mg_{8.2}Al_{2.2}Fe_{1.6})_{12}(Si_{5.8}Al_{2.2})_8O_{20}(OH)_{16}$
- Zone B: $(Mg_{7.0}Al_{2.3}Fe_{2.7})_{12}(Si_{5.7}Al_{2.3})_8O_{20}(OH)_{16}$
- Zone C: $(Mg_{5.7}Al_{2.3}Fe_{4.0})_{12}(Si_{5.7}Al_{2.3})_8O_{20}(OH)_{16}$

According to the classification of HEY (1954), the chlorites of Zone A may be termed clinochlore, those of Zones B and C are pycnochlorites. Whereas total Al is fairly constant, the Fe/(Fe+Mg) ratio ranges from 0.16 in Zone A to 0.41 in Zone C. This trend can possibly be related to the observed hematite depletion quoted above, since the iron content of the rocks and of the muscovites (RM values) is unchanged in all the zones.

Tectonic control of the metamorphism

The cause of the metamorphism in the southern part of the Stavelot - Venn Massif has often been a point of discussion. Three points of view have been developed:

- dynamic metamorphism as a result of the resistance of the Massif against the NW vergent Hercynian folding (SCHMIDT, 1956; GEUKENS, 1957);
- thermal metamorphism due to a local magmatic body (RICHTER, 1962; FOURMARIER et al., 1968);
- regional metamorphism in connection with a large-scale heating of this part of the Rhenish Mass (FRANSOLET et al., 1977).

Among these hypotheses, the first appears to fit best with the observations for the following reasons:
- there is a close relationship between the nature of the schistosity and the metamorphic zones. Whereas a non-planar and non-penetrative schistosity characteristically occurs in Zone A, it grades to a planar and penetrative type in Zone C (typological classification of RICHERT, 1974).
- SPAETH (1979) draws attention to the exceptionally strong internal deformation of the rocks along the southern border of the Stavelot Massif, which he considers "ein mächtiger Bewegungshorizont".
- Very comparable metamorphic zones can be traced along the southern margins of the other Caledonian Massifs, situated southwest of the Stavelot Massif.

BEUGNIES (1976) stressed this fact by his observation that the metamorphism in the Ardennes typically appears to be a "métamorphisme de position". Colour distributions analogous to those described in this study also occur around the other Massifs.

Conclusion

A progressive sequence which encompasses the fields of diagenesis to low-grade metamorphism was established on the basis of illite crystallinities for the Gedinnian shales surrounding the Stavelot-Venn Massif. The colour of the shales proved to be a useful tool for the mapping of low-grade metamorphic zones in the field, although the colour is believed to be more a consequence of recrystallization of hematite than to mineralogical or bulk chemical changes. Muscovite compositions indicate that the low-grade metamorphism in the southern part of the Massif is characterized by low pressure conditions and temperatures of about 415 °C. This is in good agreement with the temperature estimates of KRAMM (1973) and FRANSOLET et al. (1977) (360° to 400° for the Upper Salmian rocks) and with those of SCHREYER (400° to 450° based on spessartine garnets in the Gedinnian conglomerates).

The aluminous composition of the rocks is responsible for the Alrich muscovites, pyrophyllite and paragonite. Paragonite occurs characteristically as a mineral of the epizone.

A temperature gradient of about 115° between the northern and southern margins of the Massif is thought to be caused by strain heating which resulted from the compression of the Gedinnian formations against the Caledonian Massif.

The illite crystallinity data as well as the colours of the Upper Salmian rocks show a close correspondence with those of the neighbouring metamorphic zones of the Gedinnian, indicating that the metamorphism is a predominantly, or even exclusively, Hercynian event.

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