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Conodont alteration in Devono-Carboniferous dolomites from southern Belgium

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Abstract

Conodonts from selected Frasnian and Tournaisian dolomites in southern Belgium have Colour Alteration Index (CAI) values of 3.0–4.5 or 6.0–7.0. The textural alteration of these conodonts includes various features. Most striking are the subhedral apatite crystals that surround the conodont denticles. Furthermore, fields of apatite crystals, irregular apatite envelopes and apatite pseudomorphs after calcite occur on the conodont surfaces. The higher CAIs as well as the textural alteration are probably related to dolomitization processes. Phosphates may have been supplied by decomposition of organic matter or by ascending saline fluids.

Introduction

Colour changes in conodonts are considered to be the result of 1) carbon-fixing processes of the organic matter which is dispersed in the apatite conodont skeleton, 2) oxidation and volatilization of the fixed carbon, 3) release of water of crystallization, and 4) recrystallization of the conodont apatite. These alteration processes have been examined in detail for the first time by Epstein et al. (1977), who established a linear Conodont Colour Alteration Index (CAI) scale with a range from 1.0 to 5.0, by testing field data with laboratory heating experiments. Since then, the CAI technique has been developed and refined, with numerous applications in basin analysis and the appraisal of hydrocarbon and mineralization potential. A detailed study of the colour alteration between CAI 5.0 and 8.0 was carried out by Rejebian et al. (1987).

The morphological or textural alteration of conodonts, however, has been studied less intensively. Schönlaub et al. (1976), Flajs (1980), Rejebian et al. (1987), Burnett (1988), Königshof (1992) and Helsen (1994), among others, described the morphological alteration of conodonts from metamorphic rocks. Diagenetic apatite overgrowths and crystal enlargement on the surfaces of conodonts were reported for the first time by Pierce & Langenheim (1970), who related the textural alteration to deeper burial. Some of the effects of dolomitization on the morphological alteration of conodonts have been studied recently by Nöth (1991) and March Benlloch & De Santisteban (1993).

For the present study, dolomites and dolomitic limestones from the Frasnian Lustin and Aisemont Formations (Coen-Aubert & Lacroix 1978), probably representing a lagoonal environment, were sampled near Huy. Additionally, Tournaisian conodonts were studied from the Huccorgne Dolomite (Groessens 1986) at Ben-Ahin, from dolomitic limestones and dolomites of the upper parts of the 'Yvoir facies' and the 'Bayard facies' (Lees et al. 1985) at Mettet (M.11) and St. Aubin (St.A.7), and from massive, dolomitized limestones of a Waulsortian buildup at the type locality (Rocks G and K of Dehantschutter 1990). The location of the sampling sites is shown in Figs 1 and 2. The present paper describes and compares the CAI and textural alteration of these selected conodont faunas. Special attention is paid to prismatic overgrowths around the conodont denticles.

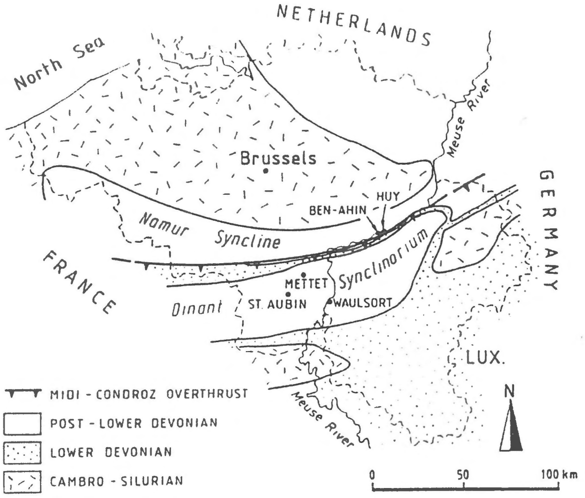


Fig. 1. Generalized map of the subsurface of Belgium, showing the geological setting of the studied outcrops of Huy, Ben-Ahin, Mettet, St. Aubin and Waulsort.

Conodont colour and textural alteration

Colour alteration

As shown by Helsen & Königshof (1994), the regional CAI in the Huy area is ranging between 3.0 and 3.5, indicating burial temperatures of 120 to 150°C. However, some of the conodonts from the dolomitized limestones have a dull lustre and/or a superficial grey patina which are probably due to oxidation and leaching of near-surface organic matter by dolomitic fluids or other oxidizing pore waters (e.g. Rejebian et al. 1987). Occasionally, the dark brown colour which corresponds to the regional CAI is recognized in broken specimens only. Apparently, the conodonts from Ben-Ahin, Mettet and St. Aubin underwent more intense interaction with oxidizing, dolomitic solutions, since most of the organic matter is volatilized. As a result, the CAI of these conodonts is anomalously high (6.0–6.5) and cannot be translated into burial temperatures (Rejebian et al. 1987). Most of the specimens have a yellow hue that may be due to trace amounts of iron. High CAIs in dolomites, as compared to non-dolomitic limestones, may also be related to differences in deformation between these rocks (Burnett 1994).

Because anomalously high CAIs of 6.0–7.0 from the Waulsortian buildups are recognized in both dolomites and massive limestones, only a part of the CAIs is believed to result from migrating dolomitizing fluids. The regional CAI in this area is 4.0–4.5, corresponding to palaeotemperatures of 190 to 245°C (Helsen & Königshof 1994).

Textural alteration of conodont denticles

The most striking morphological alteration of the examined conodonts is observed at the denticles of the ramiform elements. Apparently, these denticles are altered into hexagonal prisms (Pl. 1, Figs 1–9, Pl. 2, Figs 1, 2). Minute cracks in the crystal faces of these prisms reveal that the latter are only superficial envelopes and that the denticles are not recrystallized (Pl. 1, Figs 5, 6). Underneath the newly formed crystal faces, the original rounded teeth can be recognized. Scanning Electron Microprobe (SEM) analyses of the mineral overgrowths indicate an apatite composition. The apatite prisms described above resemble the recrystallized denticles of conodonts from metamorphic rocks (Helsen, 1994). Platy apatite crystals in between the denticles of the ramiform elements may join several denticles together, partly or completely, into narrow ridges (e.g. Pl. 1, Figs 2, 5). Occasionally, nodes of Pa elements may be altered into short subhedral crystals as well. Nearly half-spherical bumps with a diameter of approximately 4 μm, occur on the crystal face of an altered denticle (Pl. 1, Fig. 9). These structures may be the result of locally important apatite precipitation. In a later stage of alteration, the newly formed apatite prisms may become etched and corroded (Pl. 1, Figs 8, 9).

Furthermore, there is evidence for a relationship between the textural alteration and the deformation of conodonts. Figures 1 and 2 of Plate 2 show a slightly deformed *Protognathodus* Pa element, with the most prominent apatite precipitation on a denticle located at the place of deformation. This texturally altered denticle suggests a post- or syn-deformational timing for at least part of the studied alteration features. The deformation of such isolated conodont elements, which may be attributed to tectonic stress (Sintubin & Helsen in prep.), is rather uncommon in the Carboniferous of the Dinant Synclinorium.

Other examples of subhedral apatite overgrowths around conodont denticles are reported from Wisconsin (redeposited Lower Ordovician, Grether 1977: pl. 1, fig. 37), from New York and Ontario (Silurian, Kleffner 1991), from Nova Scotia and Newfoundland (Viséan Windsor and Codroy Groups, Von Bitter & Austin 1984: pls 17, 18, Von Bitter & Plint-Geberl 1982: pls 5–7), from Pakistan (Permian of

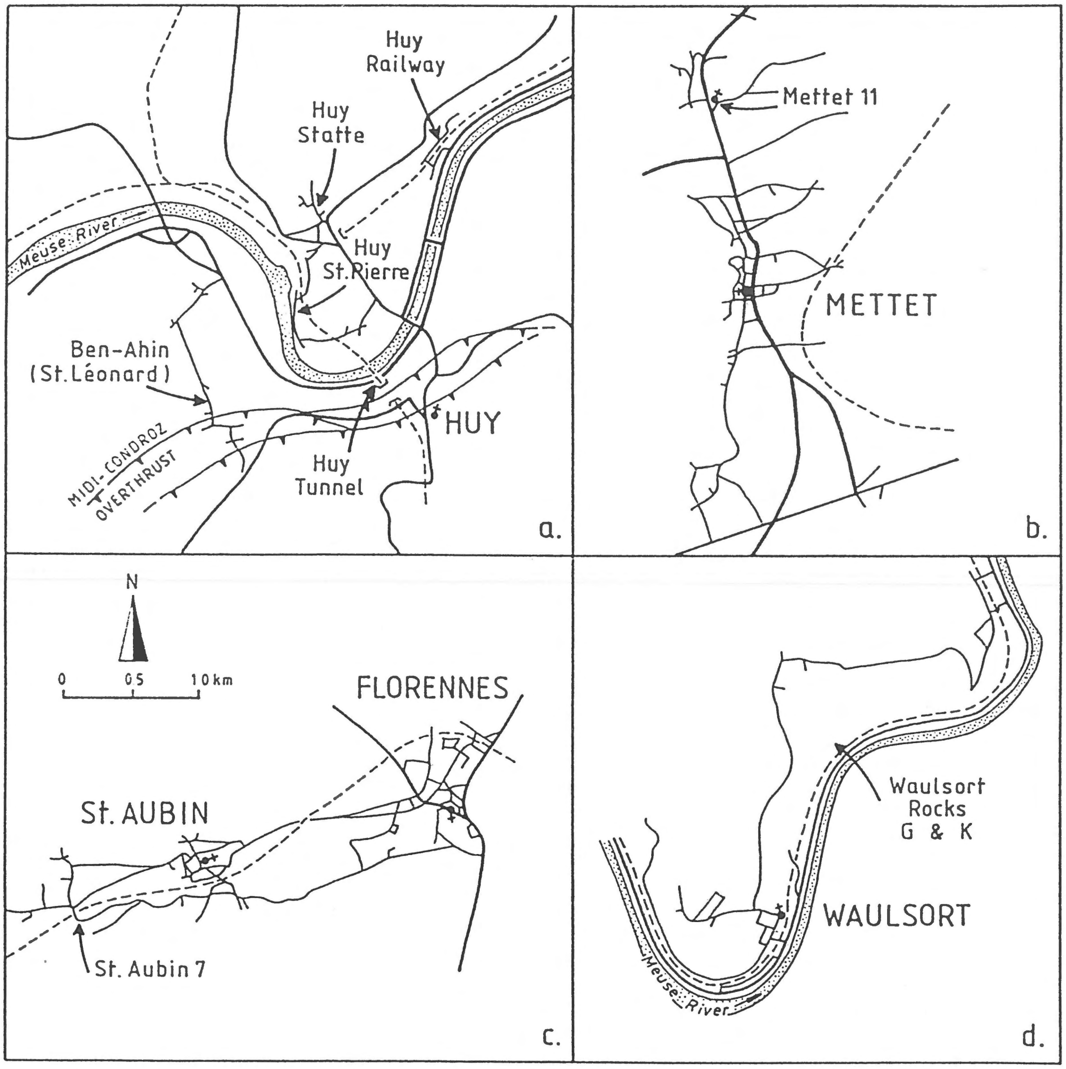


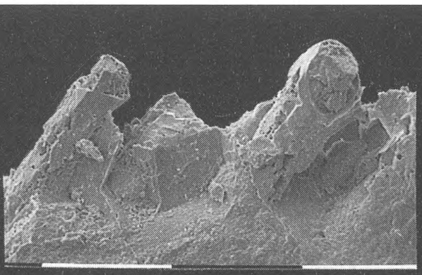
Fig. 2. Locality maps of the sampled sections: (a) Huy area, (b) Mettet, (c) St. Aubin, (d) Waulsort. Scale in (c) applies to all maps.

the Salt Range, B.R. Wardlaw, pers.comm. 1994) and from Poland (Okluscz Beds, Middle Muschelkalk, J.E. Repetski, pers.comm. 1994). Von Bitter & Austin (1984) noticed that among different genera, apatite overgrowths are chiefly present on the elements of the *Taphrognathus transatlanticus* apparatus, and that the overgrowths are best developed on the upper sides.

Subhedral and anhedral apatite envelopes and pseudomorphs

Most of the original ornamentation of the conodont denticles and the conodont platforms, including some of the smaller nodes, may be masked by apatite envelopes and scaly and platy crystals (Pl. 2, Figs 3–5). Similar apatite envelopes were observed by Alberstadt & Repetski (1989, Ordovician, Mississippi) and by Kleffner (1991, Silurian, New York and Ontario). Other examples are from the Mississippian of Spitsbergen (W. Buggisch, pers.comm. 1994), New Mexico (unpublished USGS report WMR-92–3) and Colorado (Armstrong et al. 1992). A detailed study of texturally altered conodonts from Triassic dolomites in Spain showed examples of irregular apatite envelopes (March Benlloch & De Santisteban 1993).

Probably, many of the irregular and angular structures on the surfaces of the Belgian conodonts are the result of pseudomorphism after calcite, e.g. the rhom-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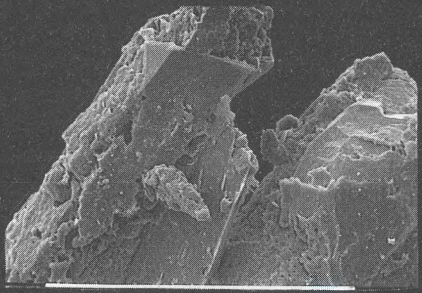
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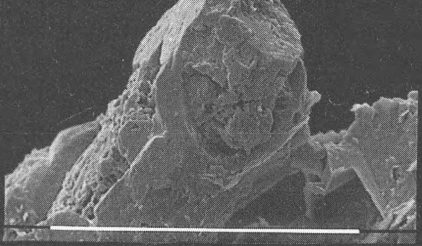
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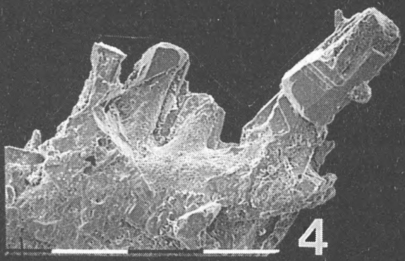
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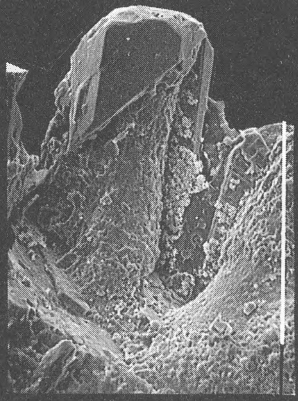
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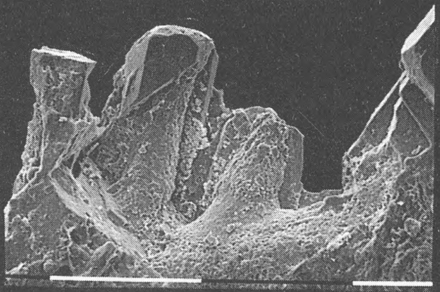
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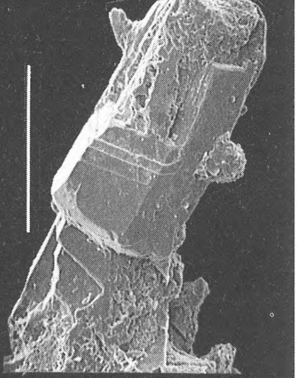
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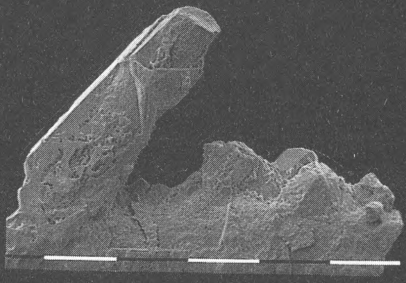
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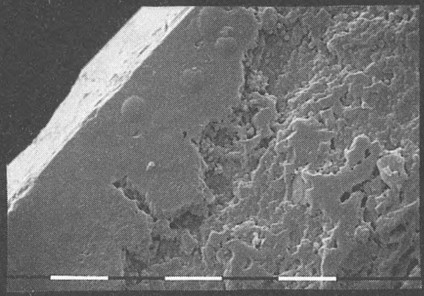
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Plate 1.

Fig. 1. Subhedral apatite envelopes of partly fused denticles of an unidentified Sc element (lateral view, *Ancyrognathus triangularis* Zone (Ziegler 1962), Frasnian, Huy-Tunnel, scale bar = 100 μm).

Figs 2, 3. Details from Fig. 1, scale bar = 100 μm.

Fig. 4. Subhedral apatite envelopes of the denticles of an unidentified Sb element (lateral view, *Ancyrognathus triangularis* Zone (Ziegler 1962), Frasnian, Huy-Tunnel, scale bar = 100 μm). Considering its orientation relative to the conodont element, the larger denticle to the right has probably been broken and grown together.

Fig. 5. Detail from Fig. 4, scale bar = 100 μm. Note the original denticle underneath the apatite coating.

Figs 6, 7. Details from Fig. 4, scale bar = 100 μm.

Fig. 8. Partly corroded subhedral apatite envelope of the denticle of an unidentified Sc element (lateral view, *Ancyrognathus triangularis* Zone (Ziegler 1962), Frasnian, Huy-Tunnel, scale bar = 100 μm).

Fig. 9. Detail from Fig. 8, scale bar = 10 μm. Note on the crystal face the half-sperical bumps which may be the result of localized apatite precipitation.

boedric apatite crystals on Pl. 2, Figs 9 and 10. When only the joints in between rhomboedric casts are filled with apatite, impressions of rhomboedric shape are observed. March Benlloch & De Santisteban (1993) concluded that this remobilization of phosphates on the conodont surfaces can produce anomalously high CAIs which are definitely not related to burial temperatures.

Crystal fields and spherical precipitations of apatite

Fields of subhedral apatite crystals are observed anywhere on the conodont surfaces (Pl. 3, Figs 1–8), but they occur most commonly on the upper sides (cf. Von Bitter & Austin 1984). Some of these fields show rhomboedric patterns which relates them to the impressions of calcite crystals and to pseudomorphism. The orientation of the crystals may be radial or random. Dense concentrations of annealed apatite crystals may result in complete overgrowth and unidentifiable specimens. Nöth (1991) recorded a continuous growth during diagenesis of newly formed apatite crystals on Triassic conodonts from Germany. This growth of apatite crystals agrees well with the observations on conodonts from contact-metamorphosed rocks by Burnett (1988).

Rounded apatite structures of less than 5 μm occasionally cover parts of the conodont surfaces. Some examples demonstrate that these ‘spherules’ can grow together in continuous envelopes (Pl. 2, Figs 6–8).

Corrosion

Besides the corrosion of newly formed crystal faces on texturally altered denticles, effects of corrosion also occur on the platforms of some of the studied specimens. The corrosion on these Pa elements is most prominent at the nodes on the platforms. This corrosion is thought to be related to the dolomitization, although the effect of artificial etching by formic acid during the sample processing cannot be excluded (Pl. 3, Fig. 9).

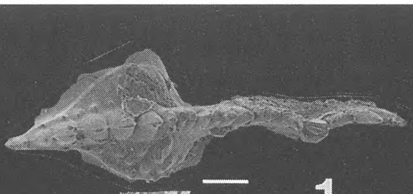
Parallel cracks

Parallel cracks in the crystal faces of some selected altered denticles and in the lateral processes of the ramiform elements, shown on Pl. 3, Fig. 10, may correspond to cleavage planes. The parallel platy structures observed on an *Ancyrodella* Pa element from Huy-Tunnel are interpreted as cleavage-related vertical cracks, which are filled with remobilized phosphates (Pl. 2, Fig. 3). Where broken denticles are grown together, kinks indicate the places of deformation, e.g. Pl. 1, Fig. 4.

Interpretation and conclusions

Textural alteration

The surfaces of conodonts from selected dolomitic rocks in southern Belgium are covered by apatite overgrowths and pseudomorphs. These overgrowths often consist of newly formed subhedral apatite crystals; most of them are enveloping the conodont denticles. The overgrowths are best developed on the upper sides, which may indicate the instability of the conodont ‘white matter’ (Sweet 1988: 14) under certain physicochemical conditions (cf. Von Bitter & Austin 1984).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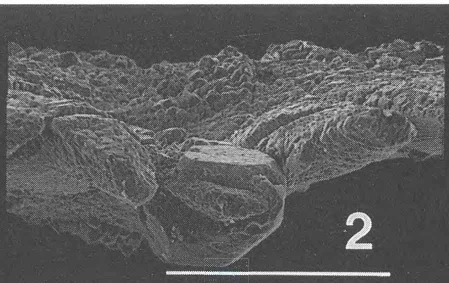
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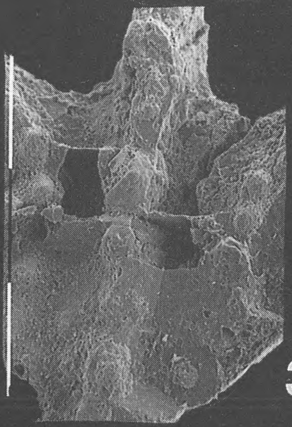
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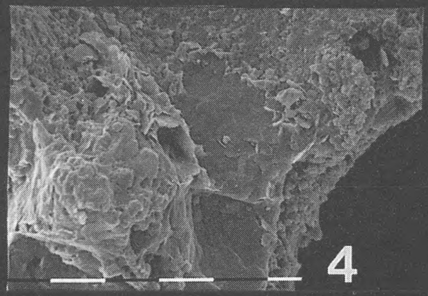
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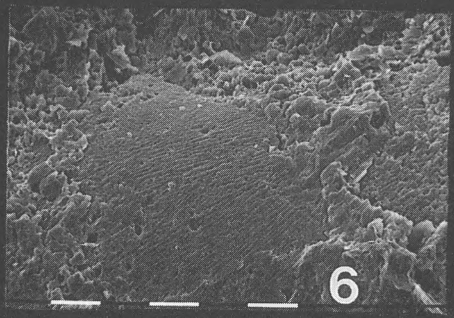
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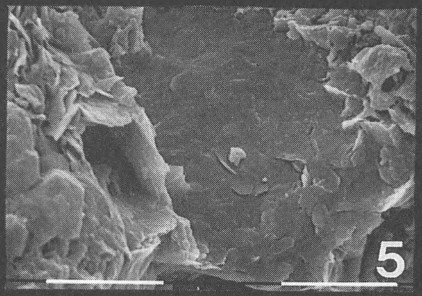
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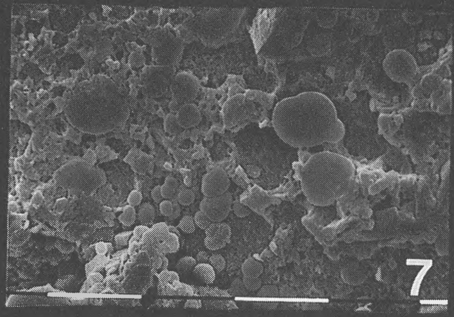
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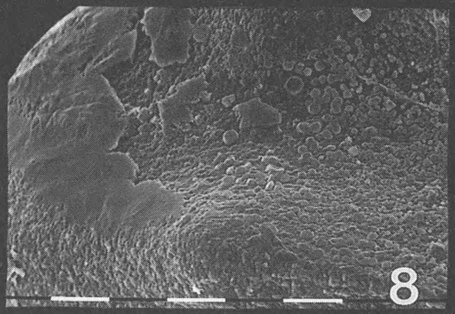
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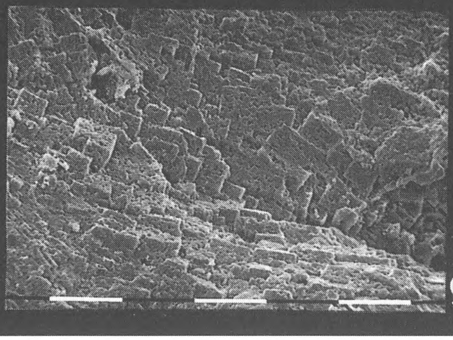
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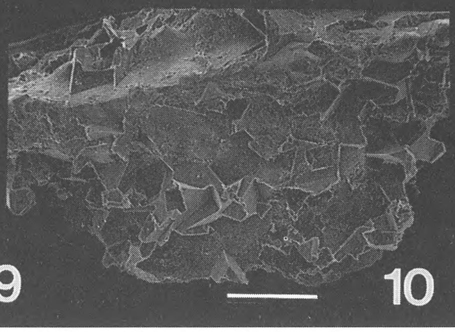
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Plate 2.

Fig. 1. Slightly deformed Pa element of *Protognathodus cordiformis* (upper view, *Polygnathus communis carina* Zone (Groessens 1971), Tournaisian, Waulsort, Rock K (Dehantschutter 1990), JD 115, scale bar = 100 μm).

Fig. 2. Detail from Fig. 1, scale bar = 100 μm. Note the apatite envelope of the denticle located at the place of deformation.

Fig. 3. Pa element of *Ancyrodella curvata*, showing parallel vertical cracks filled with apatite (upper view, *Ancyrognathus triangularis* Zone (Ziegler 1962), Frasnian, Huy-Tunnel, scale bar = 100 μm). Scaly and platy crystals cover the platform and nodes.

Figs 4, 5. Details from Fig. 3, scale bar = 10 μm.

Fig. 6. Rounded apatite structures ('spherules') on an unidentified Sb fragment (lateral view, *Polygnathus communis carina* Zone (Groessens 1971), Tournaisian, Mettet 11(2), scale bar = 10 μm)

. Fig. 7. Unidentified Sc fragment with 'spherules' of apatite (lateral view, *Polygnathus communis carina* Zone (Groessens 1971), Tournaisian, Mettet 11(3), scale bar = 10 μm).

Fig. 8. 'Spherules' of apatite coating a Pa element of *Polygnathus communis carina* (upper view, *Polygnathus communis carina* Zone (Groessens 1971), Tournaisian, Waulsort, Rock K (Dehantschutter 1990), JD 115, scale bar = 10 μm).

Fig. 9. Rhomboedric pseudomorphous apatite crystals on the free blade of a Pa element of *Polygnathus* sp. (lateral view, *Polygnathus communis carina* Zone (Groessens 1971), Tournaisian, Waulsort, Rock G (Dehantschutter 1990), JD 127, scale bar = 10 μm).

Fig. 10. Rhomboedric impressions on a Pa element of *Protognathodus cordiformis* (upper view, *Polygnathus communis carina* Zone (Groessens 1971), Tournaisian, St. Aubin 7(3), scale bar = 100 μm).

Textural alteration and CAI

Apatite overgrowths occur on conodont elements with various CAI. Besides in the Belgian samples with background CAIs of 3.0–3.5 or 4.0–4.5, textural alteration is also present on conodonts with very low CAI, e.g. samples from Nova Scotia, Newfoundland and Wisconsin (CAI 1.0) and from Pakistan and Poland (CAI 1.0–1.5). This implies that the textural alteration of conodonts can take place at an early stage of diagenesis under regional temperatures of 50° C or less. Specimens with anomalously high CAIs of 6.0–6.5 (e.g. Waulsort samples) may be texturally altered as well. However, a close relationship between this alteration and the CAI is not observed. A grey patina, frequently associated with dolomitization (e.g. Rejebian et al. 1987), is common on the texturally altered conodonts.

Textural alteration and dolomitization

More important, however, is the relationship between the conodont alteration and the lithology of the host-rock. All samples taken outside Belgium and discussed in this paper are from evaporitic sequences and dolomites, where saline pore waters could alter the conodonts through corrosion, oxidation and apatite precipitation. On the basis of our observations on material from Belgium, it seems likely that texturally altered conodonts from non-metamorphic rocks are always associated with dolomitization processes.

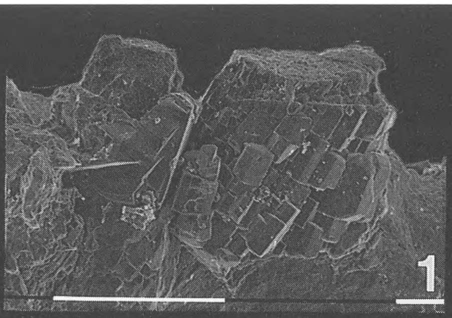
Extensive subhedral apatite envelopes probably develop preferentially in porous host-rocks, which have sufficient intergranular space. This supports the relationship between apatite precipitation and processes of replacement by dolomitization. The subhedral apatite envelope around one denticle located in the bending zone of a deformed *Protognathodus* Pa element (Pl. 2, Figs 1, 2), suggests a post- or syn-deformational textural alteration.

A preliminary, conventional cathodeluminiscence-petrographical study of several of the conodont samples reveals repeated dolomitization and dedolomitization. Thin sections of the dolomitic limestone from Huy-Tunnel showed at least five phases of calcite and dolomite crystallization.

Modes of apatite precipitation

Gaudette & Lyons (1980) summarized four possible modes of phosphate – calciumcarbonate interactions in the sediment and pore water. The direct precipitation of authigenic apatite occurs when large amounts of phosphate are added to the system, whereas amorphous calcium phosphate will precipitate only from sea-water that is supersaturated in phosphorus. A third mode, which is primarily dependent on the pH and the carbonate alkalinity, is the replacement of calcium carbonate by apatite. March Benlloch & De Santisteban (1993) concluded that for the alteration of conodonts, a fourth mode, i.e. the absorption of reactive phosphate by calcium carbonate in the presence of magnesium, is probably the most important mechanism. It appears that this absorption is enhanced in the presence of organic coatings covering the $CaCO_3$ minerals (Gaudette & Lyons 1980).

Since dolomitization requires a high, and apatite formation a low Mg^{2+}/Ca^{2+} ratio, relative to that of seawater, the two minerals probably form under differ-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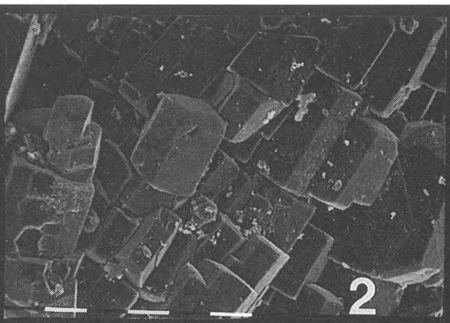
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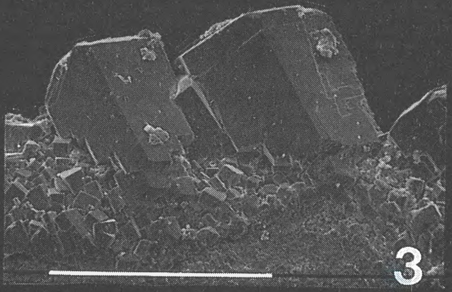
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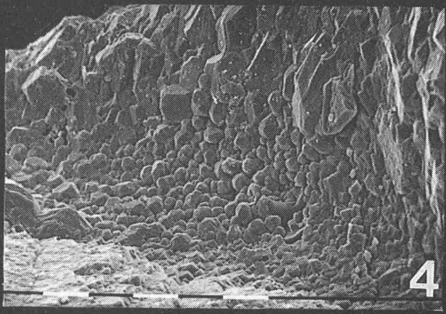
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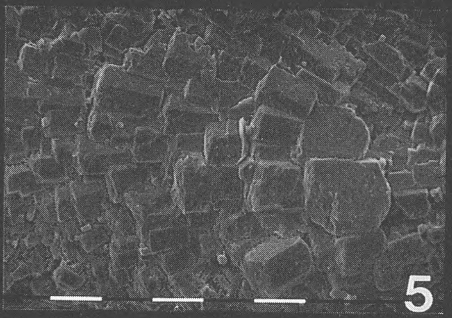
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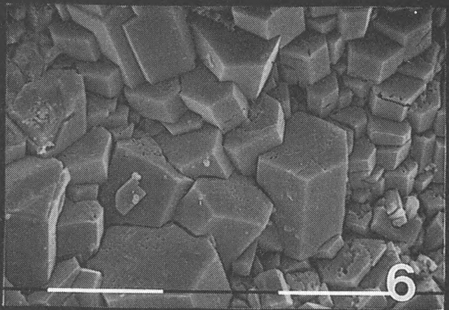
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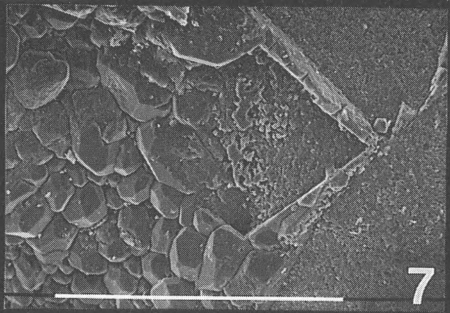
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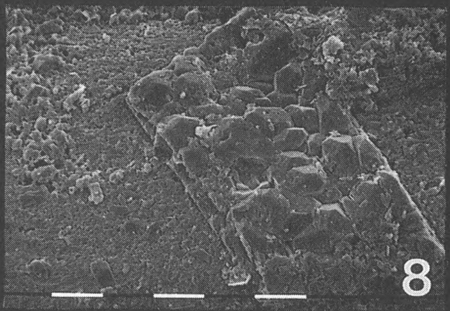
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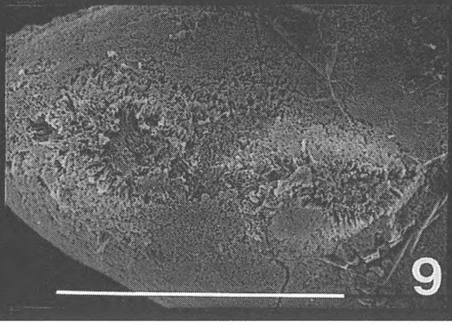
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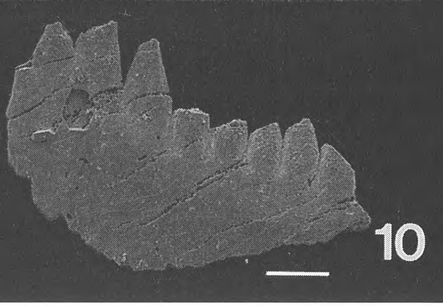
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Plate 3.

Fig. 1. Subhedral apatite crystals on a slightly deformed, unidentified S element (lateral view, *Polygnathus communis carina* Zone (Groessens 1971), Tournaisian, Ben-Ahin (St. Léonard), scale bar = 100 μm).

Fig. 2. Detail from Fig. 1, scale bar = 10 μm.

Fig. 3. Large subhedral apatite envelopes around the denticles and smaller blocky apatite crystals on the surface of an unidentified S element (lateral view, *Polygnathus communis carina* Zone (Groessens 1971), Tournaisian, Ben-Ahin (St. Léonard), scale bar = 100 μm).

Fig. 4. Blocky apatite crystals on a Pa element of *Polygnathus communis* cf. *dentatus* (upper view, *Polygnathus communis carina* Zone (Groessens 1971), Tournaisian, Waulsort, Rock G (Dehantschutter 1990), JD 127, scale bar = 10 μm).

Fig. 5. Field of apatite crystals on the surface of an unidentified S element (lateral view, *Ancyrognathus triangularis* Zone (Ziegler 1962), Frasnian, Huy-St. Pierre, scale bar = 10 μm).

Fig. 6. Subhedral apatite crystals on a Pa element of *Polygnathus mehli* (upper view, *Polygnathus communis carina* Zone (Groessens 1971), Tournaisian, Ben-Ahin (St. Léonard), scale bar = 10 μm).

Fig. 7. Apatite crystals and rhomboedric patterns on an unidentified Pb element (lateral view, *Polygnathus communis carina* Zone (Groessens 1971), Tournaisian, Waulsort, Rock K (Dehantschutter 1990), JD 115, scale bar = 100 μm).

Fig. 8. Rhomboedric field of apatite crystals on an unidentified S element (lateral view, *Polygnathus communis carina* Zone (Groessens 1971), Tournaisian, St. Aubin 7(1), scale bar = 10 μm).

Fig. 9. Corroded Pa element of *Polygnathus mehli* (lateral view, *Polygnathus communis carina* Zone (Groessens 1971), Tournaisian, Ben-Ahin (St. Léonard), scale bar = 100 μm).

Fig. 10. Fragment of free blade of Pa element of *Polygnathus* sp., showing parallel cracks which may correspond to cleavage planes (lateral view, *Polygnathus communis carina* Zone (Groessens 1971), Tournaisian, Mettet 11(3), scale bar = 100 μm).

ent physico-chemical conditions (Bentor 1980). Beside the primary deposition of apatite, replacement of calcium carbonate may thus be important. The dissolution of calcite and/or the depletion of magnesium through dolomitization decreases the Mg^{2+}/Ca^{2+} ratio and therefore facilitates the apatite precipitation (Bentor 1980). The etching of newly formed apatite envelopes on some of the conodonts, which is probably the result of dolomitic solutions, suggests a remobilization of phosphates. However, the preliminary cathodeluminiscence study shows no evidence for significant concentrations of phosphate in the carbonate rock.

Potential sources of phosphate

The increase of phosphates in the pore waters by important dissolution of conodont elements is very unlikely, since even the more fragile specimens are still preserved in the studied samples and thus were not dissolved preferentially. Moreover, it appears that in most dolomitic rocks investigated here, conodonts are too scarce to produce the necessary amount of phosphate.

In order to explain the phosphates in the Triassic carbonates of the Valencia area (Spain), March Benlloch & De Santisteban (1993) referred to Gaudette & Lyons (1980), who examined recent organic-rich shallow-water carbonates of the Bermudas. Gaudette & Lyons (1980) demonstrated that dissolved reactive phosphate, produced during the decay of organic matter, is rapidly removed, and absorbed onto $CaCO_3$ mineral grains. It is thus possible that saline dolomitizing waters, circulating in the carbonate sediment, caused a synsedimentary or early-diagenetic apatite precipitation on the surfaces of the isolated conodont elements. This is in agreement with the shallow-marine and lagoonal environments of the sampled carbonates in Belgium (e.g. Huy area) and of the evaporitic strata studied elsewhere (Kleffner 1991, Von Bitter & Austin 1984, B.R. Wardlaw, pers. comm. 1994).

However, the possibility of phosphorus supply and remobilization by ascending saline solutions cannot be excluded. In fact, in the Huy area the textural alteration of conodonts is more severe in the vicinity of fault planes of the Midi-Condroz Overthrust. These planes may have enabled saline fluids to pass through the rocks in larger quantities. The origin and migration of these fluids in the Huy area is documented by Muchez et al. (1995). Possibly, these fluids caused a secondary, late-diagenetic dolomitization of the carbonate rocks. Likewise, a number of thrust planes, recently mapped in the Waulsort area (Delcambre & Pingot 1993), may have been important in the dolomitization of the Waulsortian buildups and the conodont alteration.

Problems that still need to be investigated further include the origin of the phosphates and the timing, synsedimentary or later, of their precipitation on the conodont elements.

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