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Timing of veining and Variscan deformation in the Stavelot-Venn Massif (Belgium)

Stijn DEWAELE[1], Ariël BOVEN[1], Tom THYS[2] & Philippe MUCHEZ[2]

[1]*Department of Geology and Mineralogy, Royal Museum for Central Africa, Leuvensesteenweg 13, B-3080 Tervuren, Belgium*
[2]*Geodynamics & Geofluids Research Group, Katholieke Universiteit Leuven, Celestijnenlaan 200E, B-3001 Leuven, Belgium*

ABSTRACT: Petrographic investigation of the Grand-Halleux boreholes identified different generations of quartz veins that were formed during different periods of fluid migration in an environment that was decreasingly influenced by compressive deformation. The ^{40}Ar-^{39}Ar dating of muscovite associated with shear zones crosscutting two quartz vein generations indicated an age of 318 ± 4.7Ma, which allows determining the position of the different vein generations in relation to the Variscan deformation in the Stavelot-Venn Massif. This age also determines the approximate age for the Variscan syn-orogenic metamorphism in the Stavelot-Venn Massif.
Younger extension veins and prismatic quartz crystals in open spaces are associated with different generations of carbonates, associated with Pb-Zn-Cu mineralisation. This implies that this mineralisation did not form during Caledonian metamorphism or early Variscan orogenic metamorphism, but has a late to post-Variscan age. It could be linked to the post-orogenic expulsion of metamorphic fluids, but a link with the younger Pb-Zn mineralisation in the Variscan front zone is favoured based on the presence of high-salinity low temperature fluids.

KEYWORDS. Grand-Halleux, Ar-Ar age dating, Stavelot-Venn Massif, Pb-Zn-Cu mineralisation, paragenesis

1. Introduction

During the formation and evolution of sedimentary basins, large scale fluid migration can take place during different periods and by different processes. Research of ancient fluid systems is largely based on the study of vein systems, as the final product of the evolving fluid system. Different mineralogical and geochemical techniques can be applied to unravel the P-T-X-t evolution and history of vein related hydrothermal systems.

Metamorphic fluids have been described in the Stavelot-Venn Massif (Ferket et al., 1998; Schroyen & Muchez, 2000) and the Ardennes Allochton (Darimont, 1984; Darimont et al., 1988; Kenis, 2004; Kenis et al., 2005). Deep burial conditions and high P-T conditions before the onset of the Variscan orogeny resulted in the formation of different generations of metamorphic fluids (Darimont et al., 1988; Zhang et al., 1997; Kenis, 2004; Kenis et al., 2005). A similar scenario of metamorphic fluid evolution, but related to the Caledonian deformation, has been described for the Brabant Massif, just north of the study area. Polysulphide mineralisation along shear zones at the southern part of this massif has been related to the circulation of metamorphic fluids (Dewaele, 2004; Dewaele et al. 2004).

Mississipi-Valley-type deposits that occur in the Variscan front zone in the eastern part of Belgium were formed due to the migration of highly saline fluids (Muchez et al., 1994). The mineralisation is known by the historical exploitation of Pb and Zn minerals in the area of Plombières, La Calamine, etc, north of Eupen (Dejonghe et al., 1993; Dejonghe, 1998). The formation of this mineralisation has been explained by the migration of fluids during Mesozoic extension. The brines were formed in closed marine environments south of the Brabant Massif during the Late Palaeozoic. These fluids migrated into the Lower Palaeozoic subsurface due to their higher density (Heijlen et al., 2001). Migration of mineralising fluids has already taken place prior the Variscan deformation, as is illustrated by the formation of syn-diagenetic baryte mineralisation at Chaudfontaine during the Frasnian (Dejonghe, 1979, 1990; Dejonghe et al. 1982, 1989; Dejonghe & Boulvain, 1993) and the formation of syn-tectonic zebra-dolomites during the Dinantian (Nielsen et al., 1994, 1998). Their deep penetration allowed the fluids to survive the

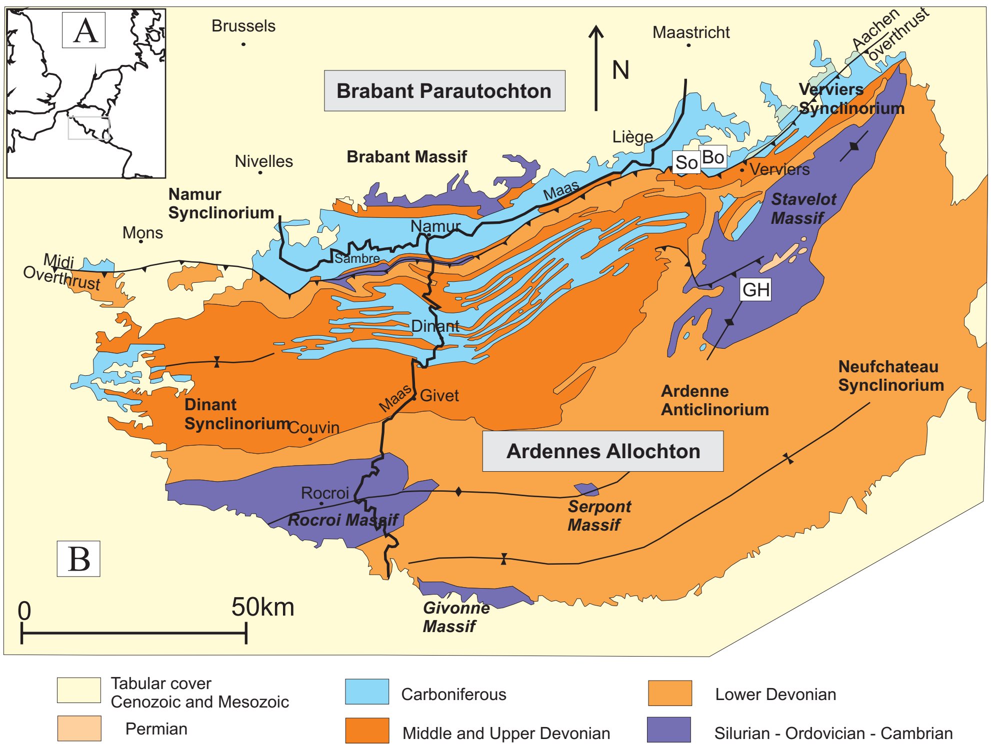

Figure 1. A. Location of the study area in its regional context. **B**. Geological map of the Ardennes Allochton (after Bultynck and Dejonghe, 2001). Location of the borehole of Grand-Halleux is indicated by GH, Bolland borehole by Bo and Soumagne borehole by So.

thin-skinned Variscan deformation. These fluids were expelled in different stages during the Mesozoic under the influence of an extensional regime and formed Pb-Zn mineralisation (Heijlen et al., 2001; Heijlen, 2002; Dewaele, 2004).

In the southern part of the Stavelot-Venn Massif of the Ardennes, three boreholes (171W276, 280 & 281) have been drilled between 1960 and 1967 at Grand-Halleux (Fig. 1) in the valleys of the Salm and a smaller attributary, near and inside the outcrop of two massifs of Devillian rocks. Other boreholes of Soumagne and Bolland (Fig. 1) are located at the Variscan deformation front. In all these boreholes, different quartz and carbonate vein generations, which can be associated with limited sulphide mineralisation, have been described (Graulich, 1967, 1975, 1977, 1980; Muchez et al., 1998).

Muchez et al. (1998) identified an uniform distribution of the palaeotemperature in veins throughout the Bolland borehole. The values obtained (~310°) are much higher than expected based on the regional metamorphism of the Dinant fold-and-thrust belt (Lünenschloss et al. 1997, 2008; Lünenschloss, 1998; Muchez et al., 1998). Due to their occurrence in the vicinity of fault systems, expulsion of metamorphic fluids from the Stavelot-Venn Massif along Variscan fault systems has been proposed as possible explanation of the temperature anomaly (Muchez et al., 1995, 1998). Two generations of fluids have been identified. Firstly, a low saline fluid has been interpreted to have a metamorphic origin. It was buffered isotopically by the Lower Devonian rocks during migration during Variscan deformation. The second type of fluid circulated after the Variscan deformation and formed carbonate minerals in extension veins.

The aim of this study is to reconstruct the paragenesis of the quartz and carbonate veins and sulphides in the Grand Halleux boreholes of the Caledonian Stavelot-Venn Massif, to date the vein systems and to determine the temporal framework of the fluid circulation in relation to the Caledonian and Variscan metamorphism, late Variscan orogenic collapse and post-Variscan extensional tectonics.

2. Geological Setting

2.1. The Stavelot-Venn Massif

The Stavelot-Venn Massif forms the northeastern part of the Ardenne Allochton (Fig. 1), which is part of the Rhenohercynian fold-and-thrust belt (Fielitz & Mansy, 1999; Franke, 2000; Oncken et al., 1999; 2000). The allochton is divided into the Dinant fold-and-thrust belt, composed of Middle Devonian to Carboniferous rocks, and the High-Ardennes slate belt, predominantly consisting of Lower Devonian rock sequences (Meilliez & Mansy, 1990; Hance et al., 1999). The High-Ardennes slate belt is subdivided in the northern Ardenne culmination that contains numerous Lower Palaeozoic basement inliers, like the Stavelot-Venn inlier, and the southern Eifel depression.

The Stavelot-Venn Massif lies in the immediate vicinity of the Variscan front complex and has been interpreted as an allochtonous nappe that has been disconnected from the basement and transported for 20 to 30 km northwestwards during Variscan thrusting on top of the Brabant parautochton (Adams & Vandenberghe, 1999), of which the Brabant Massif forms the core, confined by low-angle detachment faults. This resulted in a “thin-skinned” fold-and-thrust belt. The multiphase tectonic deformation formed a ramp-anticline, with a complex internal duplex structure of in-sequence and out-of-sequence thrust faults was formed (Fielitz, 1992; Laloux et al., 1996; Hance et al., 1999). Different structural domains can be recognised in the Stavelot-Venn Massif (Geukens, 1986, 1999). A northern domain consists mainly of rocks belonging to the Revinian Group and shows a succession of closed synclinal structures that have a N50E trend interpreted to be of Variscan origin. The trend was induced due to the presence of the Brabant Massif as a rigid bloc during the NW-oriented Variscan deformation (Hance et al., 1999; Mansy et al., 1999). The deformation at the northern margin is dominantly brittle in nature and resulted in an overprinting of earlier formed Caledonian E-W oriented structures. It should, however, be mentioned that the presence of a Caledonian cleavage is strongly debated. The southern domain, situated south of the Permian Malmedy Graben is characterized by E-W oriented tectonic structures, interpreted to be of Caledonian origin (Geukens, 1986; Piessens & Sintubin, 1997). Different E-W oriented faults of Caledonian origin are present in the southern part that have been reactivated during Variscan deformation. The Variscan deformation front developed progressively in a northwestern direction. The front reached the southern part of the Ardenne-Eifel area at the end of the Visean. During the closure of the Rhenohercynian ocean, a new foreland basin formed in front of the developing orogen. Deformation of the High-Ardennes slate belt occurred first during the Late Visean - Late Namurian (325 – 310 Ma; Fielitz & Mansy, 1999). During the late Carboniferous (~ 300 Ma), the Ardenne Allochton was thrusted northwards over the Brabant parautochton (Mielliez and Mansy, 1990).

The sedimentary rocks of the Stavelot-Venn Massif have been subdivided in the Deville Group, Revin Group and the Salm Group (Verniers et al., 2001). The oldest rocks of the massif belong to the Deville Group that forms the base of megasequence I (Verniers et al., 2001). These rocks are identified in two anticlinal structures in the southern part of the Massif, at Grand-Halleux and Ligneuville. The base of the Devillan is formed by the Grand-Halleux (Hourt) formation and consists of thick bancs of pale green quartzites, followed by greenish slates and quartzites, intercalated with purple slates of the Bellevaux Formation. The presence of Oldhamia indicates an Early to Middle Cambrian age (Malaise, 1883). The rocks of the Revin Group also belong to megasequence I and consist dominantly of darker colored slates, pelites and quartzites (Verniers et al., 2001). The Revin Group has been subdivided in the Wanne, La Venne and La Gleize Formations. Rocks of the Salm Group are considered of Ordovician age (Lamens, 1985; 1986). The lower Jalhay Formation consists of a heterogeneous assemblage of sandstones, silty shales and is situated below a hiatus and considered to be the top of Megasequence I. Megasequence II starts with purple, often silty shales of the Ottré Formation, characterised by its Fe- and Mn-rich layers (Verniers et al., 2001). Due to the onset of the Caledonian deformation in the south, Megasequence III is missing in the Ardennes inliers (Verniers et al., 2001) and a hiatus from Late Ordovician to early Devonian is identified. The Petit Tailles Formation is described, but the exact stratigraphic position is disputed (Geukens, 1999; Verniers et al., 2001). The first sediments that were deposited along the Stavelot-Venn Massif are of Late Lochkovian age and are characterised by a diachronous character due to a northwestwards transgression of the Devonian sea (Verniers et al., 2001).

2.2. The Grand-Halleux borehole

Three boreholes have been drilled at Grand-Halleux between 1960 and 1967 in the valleys of the Salm and a smaller attributary in the southern part of the Stavelot-Venn Massif, near and inside the outcrop of two massifs of Devillian rocks (Graulich, 1980). The deepest borehole (171W276 – 3225 m) was emplaced at the centre of the massif and was intended to reach older rocks than the Devillian, since the structure of the massif was interpreted to be an anticlinal “pop-up” structure, and to identify important fault structures. However, until a depth of 2360 m rocks of Devillian age were encountered followed by younger rocks of Revinian age (Franssen & Michot, 1969; Graulich, 1980). The contact between the Revinian and the Devillian is fault related, identified as the Rochelinval fault by Geukens (1986), although discussed by several authors (Franssen & Michot, 1969; Geukens, 1977; Graulich, 1980).

The Devillian rocks consist of bleu grey to light coloured quartzites that are strongly veined. These quartzites alternate with grey to green shales, locally enriched in chlorite and sericite. Often these shaly layers, less competent than the quartzites are associated with shear planes. On these planes, elongated quartz crystals, associated with muscovite and chlorite can be recognised. Revinian rocks are massif, dark coloured shales and silty quartzites. The darker colour has been explained by the presence of organic material (Franssen & Michot, 1969). Quartz and carbonate veins can be observed throughout the entire borehole. Thickness of the veins varies from less than 1 mm to several centimeters until completely mineralised brecciates zones up to 1 m. Carbonate phases mostly occur as secondary phases in the quartz veins. Sulphide mineralisation occurs as pyrite cubes disseminated in the host-rock, associated with shear planes and different vein generations. Hatert (1996) has made a very detailed study of mineralogy and geochemistry of the sulphides in the Grand-Halleux borehole.

3. Methodology

3.1. Sampling and petrography

Rocks have been sampled from the boreholes of Grand Halleux (171W276, 280 & 281), which are accessible at the Belgian Geological Survey (Royal Belgian Institute for Natural Sciences). Based on the detailed borehole descriptions of Graulich (1967, 1980), specific levels have been sampled where polysulphide mineralisation have been described. Attention was focussed on levels where the relationship between mineralisation, shear zones and quartz/carbonate veins could be observed.

Macroscopic observation of the polished slabs allowed a first characterisation of the different mineral phases and a subdivision in different vein generations, based on crosscutting and overgrowth relationships, with specific attention on the relationship with structural elements. Different carbonates were identified by using the coloration technique of Dickson (1966). This paragenesis was further worked out in detail by using transmitted and reflected light microscopy on thin and polished sections respectively. A more advanced petrographic study of the carbonate generations was carried out with a Technosyn Cold Cathodo Luminescence Model 8200MkII, operated at 16-20 kV, ~ 600 µA gun current, with a 5 mm beam width and 6.65 Pa vacuum.

3.2. ^{40}Ar-^{39}Ar dating

^{40}Ar-^{39}Ar dating was carried out on a sericite/muscovite sample taken at 1720 m depth in borehole 171W276 at Grand Halleux. It should be noted that macrocrystalline muscovite suitable for dating is very rare. The sample was handpicked and investigated for their homogeneity by microscopy, SEM analysis and XRD. Only pure, hand-picked sericite chunks were retained for the ^{40}Ar-^{39}Ar study. Minor amounts of quartz could still be found in the sample selected, but should have no effect on the age obtained.

^{40}Ar/^{39}Ar dating was carried out at the Vrije Universiteit Brussel. After measurement of the weight of the samples, they are packed in an aluminium foil prior to radiation. These samples, together with aliquots of the LP-6 biotite standard, CaF_2 and K-glass monitors, were irradiated under Cd-shielding during 3 days in channel E30 in the DG5 carrier of the BR2-reactor of the Belgian Nuclear Research Centre at Mol. The encapsulated, irradiated sample was dropped in a high-vacuum resistance oven. Step-wise heating experiments, with numerous steps at very small temperature intervals were carried out while argon measurements were made on a MAP 216 mass spectrometer operating in static mode. Depending on the amount of released gas, the isotopes are measured with a Faraday cup or an electron multiplier. During degassing of the heated sample, the released gases were purified by circulation through different getters. These getters remove other gas components and concentrate argon. Typically the instrumental ^{40}Ar-^{39}Ar set-up contains an extraction, purification and measurement part.

Ages are calculated using a J-factor determined from interpolation between 3 J-factors, obtained on aliquots of LP-6 biotite for which an age of 128.1 ± 0.2 Ma (Baksi et al., 1996) was used. All errors for the ages are given at the 2σ-level. This accounts for the analytical errors, including the variability of the neutron flux based on dosimetry measurements with a Fe-wire. Analytical details are presented in Boven et al. (2001). The uncertainty in the J-factor is ± 1 %, but excludes errors on the K and Ca correction factors.

4. Petrography and paragenesis

Cubic and framboidal pyrite is omnipresent throughout the Devillian and Revinian rocks of borehole 171W276. The size of the cubic pyrite can vary between less than 1mm to larger than 1 cm. Compaction folding and pressure shadows filled with quartz can be found associated with larger pyrite cubes. Muscovite, chloritoid, garnet, plagioclase, iron oxides and dawsonite can occur associated with pyrite (Franssen & Michot, 1969).

The quartz and carbonate veins are the scope of the study and can be observed throughout the entire borehole 171W276. Thickness of the veins varies from less than 1 mm to several centimeters until completely mineralised brecciates zones up to 1m. Carbonate phases mostly occur as secondary phases in the quartz veins. Veining, folding and faulting is more intense and veins are generally larger in the Devillian rocks than in the Revinian rocks.

Four different quartz vein (Fig. 2) generations have been distinguished based on crosscutting relationships, occurrences, morphology and orientation. Quartz generation Q0 occurs as small veinlets throughout the host-rock and in quartzite fragments that occur in breccia. This quartz generation also occurs as syntaxial overgrowth on silicified host-rocks. The individual quartz grains often have a fibrous texture and are strongly deformed, as indicated by undulating extinction, grain boundary migration and subgrain development. Quartz generation Q1 can be subdivided into two groups. The first group (Q1A) occurs as fibrous quartz veins and as fragments of these veins. The quartz crystals have a pronounced length-width ratio (ratio 12:1) and show signs of an intense deformation. In some of the veins, the

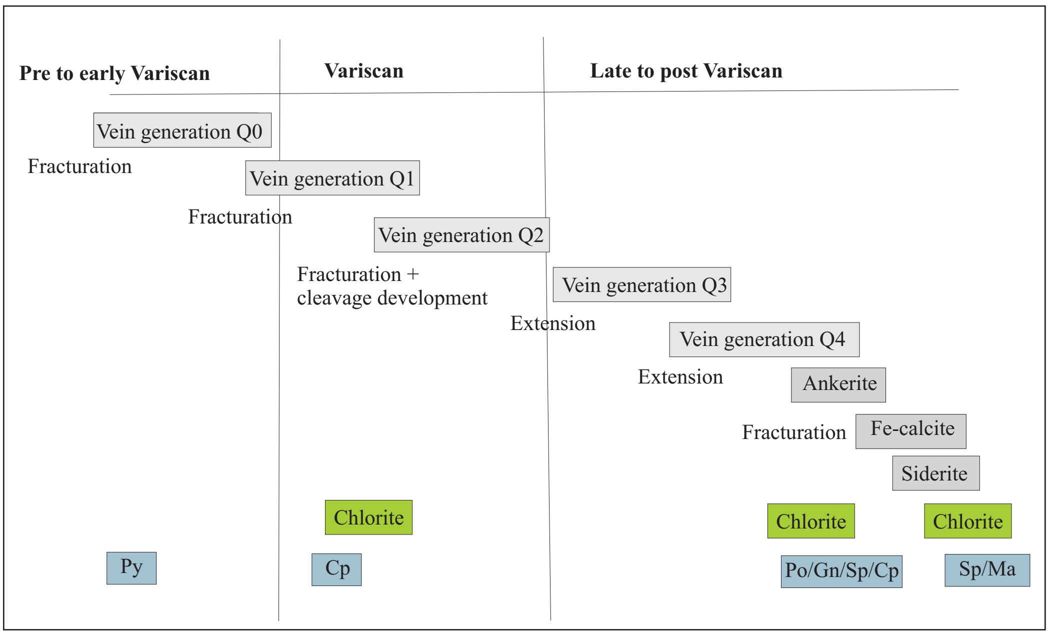

Figure 2. Paragenesis indicating different vein generations and mineralisation in the Grand-Halleux borehole. Py : pyrite, Po: pyrrhotite; Gn: galena; Sp: sphalerite; Ma: marcasite and Cp: chalcopyrite

quartz crystals in the central part of a vein bend over to a mutual shear plane. The quartz crystals in this type of veins interlock very closely, which could indicate strong grain boundary migration. In addition, strong undulating extinction and subgrain development can be observed. These veins have been fractured and carbonate crystals precipitated parallel to the quartz fibers. Quartz veins of group Q1B (Fig. 3A) are very similar to type Q1A, but consist more of saccharoidal, equidimensional quartz crystals. Q1 (and Q0) veins are crosscut by shear and fracture planes with muscovite and chlorite. They also occur in the alteration zones surrounding host rock fragments enclosed in the quartz veins. Quartz generation Q2 also consists of the two groups (Q2A and Q2B) that are strongly related since they form a continuous phase that ends in open spaces (Fig. 3A). Distinction can be made based on their appearance during microscopic observation. Q2A contains "dirty" quartz crystals due to the presence of numerous fluid inclusions, while Q2B quartz shows trails of very small fluid inclusions. The quartz crystals show slightly undulating extinction and regular geometric grain contacts. No subgrain development nor grain boundary migration can be observed, which could imply another deformation history of the previous quartz generations. Quartz generation Q3 occurs as prismatic crystals on the walls of cavities in veins and occur as overgrowths on the previous quartz generations. The quartz crystals are milky white at their base and become more transparent to the top of the prism. A mottled appearance is possible due to the presence of clusters of fluid inclusions. The quartz crystals show a slightly undulating extinction. This quartz generation occurs associated with sulphide mineralisation (Fig. 3B). Quartz generation Q4 is considered to form a continuous phase with the previous generation Q3, but can be distinguished due its clear association with extensional veins, often associated with open spaces. The crystals are sheet-like to fibrous and grow from border to border in small veins or as prismatic crystals in open spaces. The quartz crystals show uniform extinction. Small cracks form the only indication of some deformation.

Carbonate minerals occur in small veinlets throughout the host-rock and quartz veins and as secondary infill in the fractured veins and in the extensional quartz veins. Ankerite, Fe-rich calcite and siderite can be identified (Fig. 3A+B). Pink ankerite occurs as infill in quartz veins. Ankerite is often associated with sulphide mineralization. In open spaces or cracks ankerite continues into euhedral saddle dolomite, often associated with an overgrowth of siderite. The formation of the saddle dolomites is followed by the precipitation of sulphide minerals and chlorite. Siderite also occurs at the margin of the carbonate veins at the contact with the host-rocks. Fe-rich calcite can be found together with ankerite filling extensional veins. Calcite postdates ankerite since it occurs in cracks in the ankerite and since it replaces ankerite in extensional quartz veins. A clear syngenetic relationship between sulphide mineralisation and carbonate formation can be identified. A second type of chlorite occurs as vermicular inclusions in quartz and dolomite, along open fractures and as fan-like precipitates on dolomites in open cracks.

A first sulphide generation consists of chalcopyrite and is associated with the shear planes (Fig. 2). No quartz is associated with this sulphide generation. The second and largest sulphide mineralisation consists of massive veins, fillings in cavities and sporadic crystals in veins. The sulphide minerals in these veins consist of galena, sphalerite, pyrrhotite, arsenopyrite, chalcopyrite, pyrite and marcasite. The majority of the sulphides occurs in existing quartz veins that have been fractured or contain open spaces. In some samples, a first stage of sulphide mineralisation formed prior to carbonate formation. Quartz generation Q3 is overgrown and replaced by galena and pyrrhotite, followed by siderite and ankerite veins associated by sulphide mineralisation. These sulphides occur associated with pink, iron-rich dolomite, white calcite and a late siderite phase that are the infill of deformed and fractured quartz veins or of open spaces. The third group of sulphide mineralisation occurs as open space fillings associated with rhombohedral saddle dolomite at the borders. The saddle dolomites are often covered with a thin layer of siderite, followed by marcasite and black coloured sphalerite. In addition to the sulphides, chlorite occurs in this dolomite.

5. Ar-Ar dating

$^{40}Ar/^{39}Ar$ dating was carried out on a sericite/muscovite sample taken at 1720 m depth from the 171W276 borehole at Grand Halleux (Fig. 4, Table 1). The sample was selected from a muscovite/sericite shear zone crosscutting vein generation Q1 (and Q0), which predates the polysulphide mineralisation. The chlorite-muscovite slickenfibers on these cleavage planes formed during cleavage-parallel shear. Sericite/muscovite is closed for argon loss at a temperature of ~ 350°C. This temperature corresponds to the presumed temperature of metamorphism in the Stavelot-Venn Massif (Schreyer, 1975; Fransolet & Kramm, 1983) and to the measured homogenisation temperatures in presumed metamorphic vein systems in the Stavelot-Venn Massif

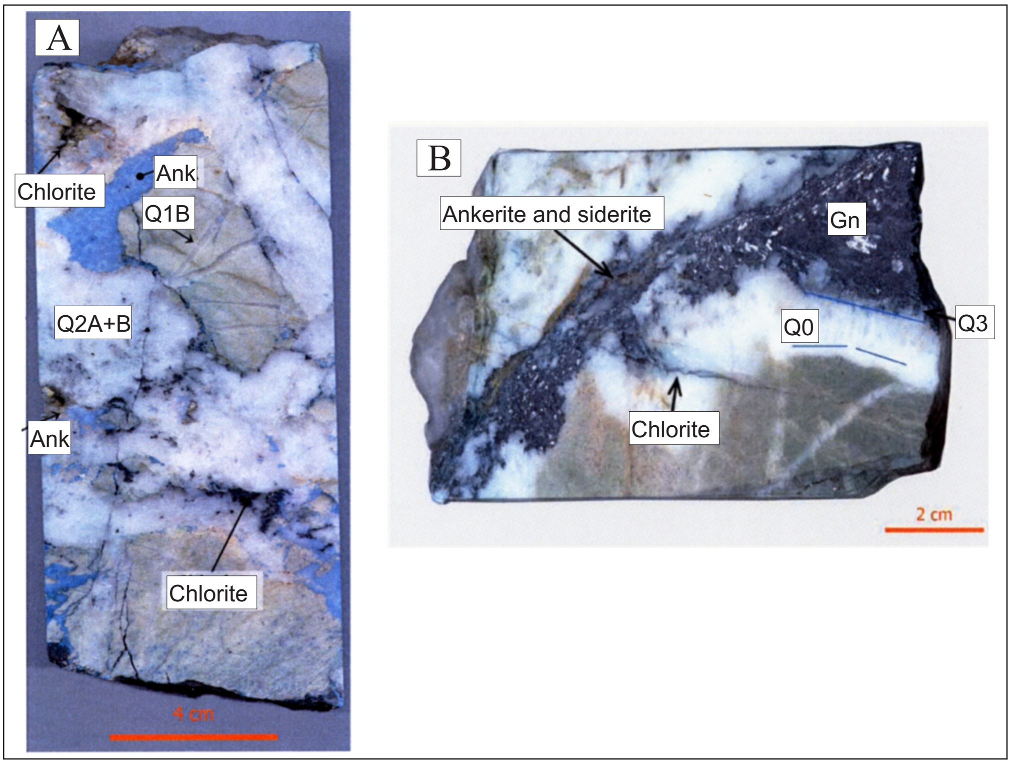

Figure 3. A. Sample from 1593 m depth of borehole 171W276 at Grand-Halleux, indicating different quartz and carbonate generations. **B.** Sample from 2253 m depth of borehole 171W276 at Grand-Halleux, indicating relationship between galena mineralization (Gn) and different quartz (Q0, Q1B, Q2A+B, Q3) and carbonate vein generations (Ank: ankerite).

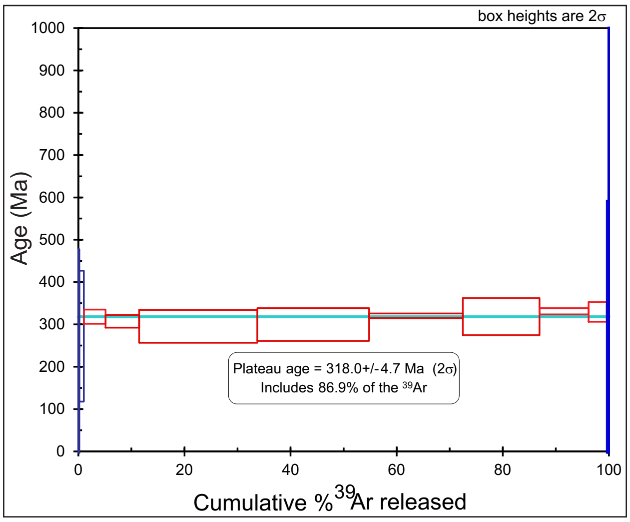

Figure 4. ^{40}Ar-^{39}Ar profile of sericite/muscovite sample taken at 1720 m depth from the 171W276 borehole at Grand Halleux. Red area has been used to calculate the plateau age.

(Schroyen & Muchez, 2000). This indicates that the muscovite/sericite grew close to its closing temperature.

The muscovite sample of Grand-Halleux yields a plateau age of 318.0 ± 4.7 Ma, which accounts for 87% of the released ^{39}Ar (Fig. 4). This age falls in the Late Carboniferous and indicates a Bashkirian age (Gradstein & Ogg, 1996). Although only one sample has been dated and the age should therefore be treated with care, it is related to an important period of deformation, cleavage development and the circulation of fluids along these structures.

6. Discussion

6.1. Formation history of Grand Halleux vein system

Different quartz and carbonate vein generations have been identified before in the Grand Halleux boreholes. The observed difference in intensity in veining between the Devillian and the Revinian rocks has been explained by a different deformation histories or the difference in lithology (Graulich, 1980). In this study, four quartz vein generations (Q1 to Q4) have been identified based on crosscutting relationships, occurrence, morphology and orientation. The absolute $^{40}Ar/^{39}Ar$ age of 318.0 ± 4.7 Ma allows to put the different quartz vein generation in more constraint temporal framework. The early Q0 quartz generation could be of Caledonian or early Variscan age. Quartz generation Q1 contains fibrous and saccharoidal crystals that show intense deformation and are crosscut by shear planes and fractures with muscovite and chlorite, like the dated sample. The quartz veins from type Q1 could therefore have a Variscan age. After quartz generation Q1 and associated shearing, multiple fracturation phases followed, characterised by a new generation of quartz veins (Q2 and Q3) that underwent less deformation. From quartz generation Q2B onwards, veins with open spaces occur. Generation Q3 is characterised by prismatic quartz crystals. This vein generation could still have formed during a late stage of the Variscan deformation, since the quartz crystals show slight indication of deformation. Quartz generation Q4 occurs as open extension veins, showing little evidence of deformation. This vein generation therefore likely formed during a late stage of the Variscan deformation or has even a post-Variscan age. Carbonates precipitated for the first time in these extension veins. Fluid migration along this system resulted in the formation of polysulphide mineralisation associated with different generations of carbonates. A first pulse of sulphide formation resulted in the precipitation of pyrrhotite, galena and sphalerite. A later pulse resulted in the formation of the pyrite, chalcopyrite and arsenopyrite. Siderite and ankerite are the earliest carbonate phases, followed by Fe-rich calcite. Ankerite can be overgrown by a second siderite generation.

6.2. Deformation and metamorphism in the Ardennes Allochton

The difference in evolution of the degree of metamorphism between the Lower Palaeozoic Massifs and their later cover in the Ardenne Allochton indicate that metamorphism developed during different periods (von Winterfeld, 1994; Helsen, 1995; Fielitz, 1992; Fielitz & Mansy, 1999). Within the regional tectono-metamorphic history of the Ardennes Allochton, a distinction can be made between a Pre-Variscan- (Caledonian), a pre-orogenic Variscan and a Variscan syn-orogenic metamorphism.

A metamorphic contrast can be found in the northeastern part of Stavelot–Venn Massif between the diagenetic Lochkovian-rocks that cover anchizonal Cambrian-Ordovician strata. The presence of the Caledonian metamorphism has been demonstrated by Ferket et al. (1998) based on the study of Caledonian vein fragments in the Lower Devonian conglomerates. The preservation of acritarches indicates that the Caledonian metamorphic influence was only of low degree (Bless et al., 1990). Indications for Caledonian metamorphism in the southern part are a matter of discussion. The favored interpretation is that the Caledonian metamorphism was completely overprinted by Variscan metamorphism (Schroyen, 2000).

The second phase of metamorphism is a pre-orogenic Variscan metamorphism. A zone of epizonal metamorphism can be found at the southern flanks of the Lower Palaeozoic inliers, 400-450°C and 100-300 MPa for the southern part of the Stavelot-Venn Massif and 500°C and 200-300 MPa for the southern margin of the Serpont and Rocroi Massifs. This zone affects both the oldest Cambrian-Ordovician and the Lower Devonian rocks, while the younger strata of Middle Devonian to Upper Carboniferous age show a much lower degree of metamorphism, implying burial metamorphism (Fielitz & Mansy, 1999; Helsen, 1995). Also the decrease in metamorphic degree from south to north corresponds with a difference of burial depth, since the sedimentary wedge developed in this direction. Local fluctuations in metamorphic degree are often due to the presence of a shear zone that bring older rocks to the surface (Han et al., 2003). This metamorphism

Temp (°C)	$^{40}Ar/^{39}Ar$	$^{37}Ar/^{39}Ar$	$^{36}Ar/^{39}Ar$ (10^{-3})	$^{38}Ar/^{39}Ar$ (10^{-3})	$^{37}Ar/^{38}Ar$	^{39}Ar Cum (%)	$^{40}Ar^*$ (%)	$^{40}Ar^*/^{39}Ar$	$^{40}Ar^*/^{37}Ar$	Apparent Age ± 2 σ (Ma)		
Sample GH, weight = 1.6 mg, 1 chip, J = 0.972 ± 0.003												
650	1.03	0.02	0.00	0.02	0.92	0.01	0.01	0.68	38.97	115.04	±	364.78
750	1.83	0.00	0.00	0.01	0.30	0.04	0.06	1.68	441.87	272.13	±	154.69
800	2.01	0.01	0.00	0.01	0.70	0.19	0.37	1.98	231.41	318.18	±	16.67
850	1.92	0.00	0.00	0.01	0.04	0.29	0.55	1.91	3711.91	307.31	±	15.06
925	1.83	0.00	0.00	0.01	0.06	1.02	1.87	1.83	2501.64	295.36	±	38.86
975	1.86	0.00	0.00	0.01	0.07	0.96	1.79	1.86	1998.13	299.63	±	38.61
1025	2.00	0.00	0.00	0.01	0.26	0.81	1.62	2.00	636.79	320.28	±	5.37
1100	1.99	0.01	0.00	0.01	0.45	0.66	1.31	1.99	351.27	318.41	±	43.88
1150	2.08	0.02	0.00	0.01	1.29	0.43	0.88	2.07	133.95	330.96	±	7.33
1225	2.08	0.05	0.00	0.01	4.12	0.16	0.32	2.06	44.00	329.57	±	23.45
1300	1.75	0.36	0.00	0.02	19.97	0.01	0.02	1.56	4.34	254.65	±	337.88
1425	1.42	0.23	0.00	0.02	12.84	0.00	0.00	0.68	2.91	114.83	±	1466.99
1600	9.41	1.01	0.01	0.01	134.82	0.00	0.01	7.29	7.23	966.41	±	1366.23

Table 1. ^{40}Ar-^{39}Ar dating of sericite/muscovite at 1720 m in the Grand-Halleux borehole. Data indicated in red have been used to calculate the plateau age.

has been identified to be earlier than the Variscan deformation (Beugnies, 1986; Fielitz & Mansy, 1999; Fransolet & Kramm, 1983). The peak in thermal evolution took place prior to cleavage development and compressive deformation and can be linked to pre-orogenic rift development of the Rhenohercynian basin until the Carboniferous (Fielitz & Mansy, 1999). The maximum age possible for this metamorphism is indicated by the age of the magmatic intrusions in the Stavelot-Venn and Rocroi Massifs that have been submitted to metamorphism and cleavage development (373 Ma, Fielitz & Mansy, 1999; Goffette et al., 1991; McKerrow et al., 2000; 381 Ma, Kramm & Buhl, 1985). The minimum age is determined by the recrystallisation of the phyllites of Lower Lochkovian age, dated between 326 ± 7 Ma (Picqué et al., 1984) and 312-308 Ma (Kramm et al., 1985). The pre-Variscan orogenic metamorphism in the epizonal area of the Ardennes Allochton is considered to be associated with the fluid migration and the formation of the different vein generations in the area, since their structural relationship with the Variscan cleavage development favors a pre- to early synkinematic formation (Fielitz & Mansy, 1999; Kenis, 2004; Schroyen & Muchez, 2000). During burial, fluids were expelled from compartments with high -often supra-lithostatic- fluid pressures, resulting in hydraulic fracturating (Darimont et al., 1988; Kenis, 2004; Kenis et al., 2005), as is illustrated the growth of metamorphic porphyroblasts at the southern part of the Stavelot-Venn Massif (Kramm & Buhl, 1985; Fransolet & Kramm, 1983).

A Variscan syn-orogenic metamorphism is mostly found in the foreland of the Variscan front and the central part of the Dinant synclinorium. This latter zone is characterised by a higher temperature regime, i.e. between anchizone-epizone (≈300°C). This is explained by the additional burial due to infill of eroded Upper Carboniferous sediments, in combination with additional tectonic loading of nappes during the Variscan deformation (Helsen, 1995; Han et al., 2003). The surrounded areas are only characterised by a diagenetic grade. At the eastern margin of the Stavelot-Venn Massif, area of Konzen, a burial metamorphism overprinted by shear zone has been recognized and explained due to the brittle-ductile continuation of the Xhoris fault in northeastern direction. This is locally identified by a penetrative, oblique shear zone deformation, thrust faults and by an epizonal and retrograde metamorphism (Fielitz, 1992).

The age obtained at the Grand-Halleux borehole is related to cleavage development and circulation of metamorphic fluids along these planes. Similar observations have been obtained by Nierhoff (1994) and Glasmacher et al. (2001). Palaeozoic rocks in the footwall of the Aachen thrust are affected by compressive Variscan deformation which was dated by Nierhoff (1994) to have occurred between 336 and 300 Ma. Glasmacher et al. (2001) obtained an Ar–Ar age of 314 ± 7 Ma for Variscan cleavage formation and a minimum age of 300 ± 8 Ma for a later hydrothermal fluid flow event. They envisaged a flux of hot metamorphic fluids expelled from the epizonal metamorphic domains of the Stavelot–Venn massif, focused along major thrust faults of the Variscan front zone such as the Aachen thrust.

6.3. Origin of Pb-Zn-Cu mineralisation at Grand-Halleux

The main part of the Pb-Zn-Cu mineralisation observed in the boreholes at Grand-Halleux occurs associated with different carbonate generations which precipitated in extension veins, postdating the Q0 to Q2 vein generation. The origin and timing of this Pb-Zn-Cu mineralisation is difficult to constrain. Copper mineralisation have been reported in the Stavelot-Venn Massif and are thought to have formed at the end of the Variscan metamorphism (Michot, 1954-1955; de Bethune & Fransolet, 1986; Hatert, 1996; 2003; 2005). Syn-metamorphic, orogenic polysulphide mineralisation has also been described by Piessens et al. (2002) and Dewaele et al. (2004) at the southern border of the Brabant Massif. However, this mineralisation is of Caledonian age and occurs along a major shear zone. The maximum possible age of the Pb-Zn-Cu sulphides at Grand Halleux is late Variscan and from structural relationships a syn-orogenic origin can be excluded. Major fluid migration could occur during such transition of a compressional to an extensional regime, resulting in mineralisation (Sibson, 2004; Cox, 2010; Van Noten et al., 2011). In this scenario, fluid inclusions in the carbonates most likely would have a high temperature and low salinity in the Variscides of the Ardennes (Muchez et al., 1998). A second possibility is that Pb-Zn-Cu mineralisation largely post-dates the Variscan and formed during the Mesozoic such as the Zn-Pb deposits at the Variscan foreland (Dejonghe, 1998). These mineralising fluids typically have a high salinity (Muchez et al., 1994; Heijlen et al., 2001). These fluids have been identified in veins in the Lower Devonian of the Bolland borehole (Muchez et al., 1998) and in the Caledonian rocks of the Brabant Massif (Dewaele et al., 2004). Thys (2006) performed a microthermometric study of fluid inclusions in sphalerite and in iron-rich dolomite related to the polysulphide mineralisation in the Grand Halleux borehole. Primary inclusions in dolomite have an H_2O-NaCl-$CaCl_2$ composition and a salinity between 16.5 and 22.6 eq. wt.% NaCl. The homogenisation temperatures vary between 90 to 103°C. Secondary fluid inclusions have been measured in the sphalerite. Homogenisation temperatures range between 90 and 100°C and the salinity is around 22 eq. wt% NaCl. Therefore, the formation of the Pb-Zn-Cu mineralisation (and associated carbonates) at Grand-Halleux seems to be related to the circulation of a high-salinity, low temperature H_2O-NaCl-$CaCl_2$ fluid.

This would imply that the Grand-Halleux mineral occurrence precipitated from a fluid comparable to the fluid that formed the “Mississipi-Valley-type” deposits at the Variscan front zone in the eastern part of Belgium, rather than being comparable to the Early Palaeozoic mineralisation described in the Brabant Massif (Dewaele et al., 2004). The formation of the Pb-Zn mineralisation in the Variscan front zone has been explained by the migration of fluids during Mesozoic extension (Heijlen et al., 2001; Heijlen, 2002). Brines that were formed in closed marine environments during the Late Palaeozoic migrated into the Lower Palaeozoic subsurface due to their higher density. Their deep penetration allowed the fluids to survive the thin-skinned Variscan deformation. Fluids were expelled in different stages during the Mesozoic under the influence of an extensional regime and formed Pb-Zn mineralisation.

7. Conclusion

Petrographic investigation of the Grand-Halleux boreholes identified different generations of quartz veins that formed during fluid migration in an environment that was decreasingly influenced by compressive deformation. The older Q0 and Q1 quartz veins predate cleavage development, since they are crosscut by muscovite/chlorite fibers formed during cleavage-parallel shear. Quartz vein generation Q2 has been less deformed. Younger quartz vein generations (Q3 and Q4) formed as extension veins. In these open fractures, fluids circulated that caused sulphide mineralization associated with different carbonate generations.

The Ar-Ar dating of muscovite associated with shear zones crosscutting quartz vein generation Q1 resulted in an age of 318 ± 4.7 Ma, which allows to constrain the timing of the different vein generations in the Stavelot-Venn Massif. Vein generation Q1 must therefore have an early Variscan age. The 318 ± 4.7 Ma age gives also a time constraint (minimum age) for the Variscan syn-orogenic metamorphism. Multiple fracturing phases followed, characterised by quartz veins that show less deformation, which could imply a formation during a late stage of the Variscan deformation or be of post-Variscan origin.

Fluid migration along extension veins and cavities in the boreholes of Grand-Halleux resulted in the formation of polysulphide mineralisations associated with different generations of carbonates. This mineralisation has also a late to post-Variscan origin. The Pb-Zn-Cu mineralisation can be related to Mesozoic extensional tectonics, which resulted in Mississippi Valley-type deposits near the Variscan front.

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