Mineralized veins resulting from fluid flow in decollement zones of the Sicilian prism evidence from fluid inclusions

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Mineralized veins resulting from fluid flow in decollement zones of the Sicilian prism : evidence from fluid inclusions

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Key words. – Accretionary prism, Décollement, Mineralized veins, Fluid inclusions, Transient fluid flow, Sicily.

Abstract. – Imbrications took place at the frontal part of the Sicilian accretionary prism during the Plio-Quaternary.The structural setting shows a superposition of southward directed thrust-sheets bounded by two décollement levels. Syntectonic veins were sampled in thrust fault zones located at the frontal part of the prism. In the lower thrust fault zone, hydraulic breccias include quartz and calcite veins. The initial crystallisation fluids have been entrapped in these veins and preserved as different generations of fluid inclusions. Primary intracrystalline aqueous fluid inclusions are composed of low salinity water (near 0.8 wt % equ.NaCl) depleted in gas and display homogenisation temperatures increasing from quartz to calcite to values up to 200°C. In calcite veins, hydrocarbon fluid inclusions containing light aliphatic oil indicate the occurrence of organic matter in the tectonic wedge with a thermal evolution comprised within the oil window.

The pressure required is the lithostatic pressure, in agreement with the conditions implied by the tectonic setting accounting for the formation of hydraulic breccias. It ranges from 140°C-0.8kbar for quartz up to 235°C-1kbar for calcite deposition. Intersection of oil and aqueous solution isochores lead to comparable values. In addition the occurrence of pyrobitumen particles in the veins indicates that the fluids reached temperatures higher than 150°C. These high temperature fluids are localized in the lower fault zone. In the upper fault zone, syntectonic calcite veins only display low temperature fluid inclusions. The results of X-ray studies on the foliated host-rock of the veins provide evidence for a highly localized incipient thermal transformation of the clay minerals (I/S of R_3 type) at the contact with the hydraulic breccia, while the overall mass of the surrounding shales remained unaffected (I/S of R_0 type).

The strict localization of the veins in the fault zones suggest that dewatering of the Sicilian wedge occurred along high-permeability décollements and faults. In the lower fault zone, warm fluid circulation of fresh water was responsible for the hydraulic fracturing and the subsequent vein formation restricted to the basal décollement, where a thermal desequilibrium occurred between the fluids and the shale matrix. This may be interpreted as the result of large-scale advection of fluids arising from the deeper internal zone of the prism and flowing along the basal décollement.

Veines minéralisées : indices de paléo-circulations de fluides dans les décollements du prisme sicilien : données des inclusions fluides

Mots clés. – Prisme d'accrétion, Décollement, Veines minéralisées, Inclusions fluides, Circulations transitoires de fluides chauds, Sicile.

Résumé. – La zone frontale du prisme sicilien présente des veines minéralisées syntectoniques localisées dans les zones de chevauchement (mélange varicolore). Les brèches hydrauliques, échantillonnées dans le chevauchement basal, sont formées de veines de quartz et de calcite. Au moment de la cristallisation, les fluides ont été piégés en différentes générations d'inclusions. Les inclusions aqueuses primaires présentent une faible salinité (0,8 wt % NaCl) avec des températures d'homogénéisation croissantes du quartz à la calcite et atteignant 200°C. Dans les veines de calcite, des inclusions d'hydrocarbures légers attestent de la présence dans le fluide de matière organique au stade de maturation de la fenêtre à huile.

Le calcul des conditions P-T, à partir de l'évolution des isochores, en accord avec la pression lithostatique minimale nécessaire à la formation des brèches hydrauliques, conduit à des valeurs de 140°C dans le quartz à 235°C dans la calcite pour des pressions de 0,8 à 1 kb. Le recoupement de l'isochore des hydrocarbures inclus avec celui des solutions aqueuses amène à des valeurs comparables. De plus, des particules de pyrobitumes, associées aux veines de calcite, indiquent qu'elles ont atteint une température d'au moins 150°C (dans le cas d'une maturation de type « bassin sédimentaire »). Les fluides chauds ainsi caractérisés sont localisés dans la zone de chevauchement basal (mélange varicolore inférieur), on ne les retrouve pas dans les veines échantillonnées dans les zones de chevauchement plus superficielles (mélange varicolore supérieur). Les spectres X des argiles du mélange encaissant des brèches hydrauliques présentent des indices de transformation thermique des minéraux argileux (I/S de type R_3) situés uniquement au contact de la brèche.

La stricte localisation des veines au niveau des zones de failles suggère que l'expulsion des fluides, lors de la déformation au haut du prisme sicilien, s'est effectuée le long des zones de forte perméabilité (failles et décollements). Des fluides chauds et très peu salés ont circulé dans la zone de chevauchement basal, connectée au décollement inférieur du prisme, et provoqué la formation des brèches hydrauliques. Le déséquilibre thermique entre l'encaissant des brèches et les fluides piégés suggère que ces fluides n'ont pas été chauffés *in situ*, mais ont migré, le long du décollement, depuis des zones plus internes et plus profondes du prisme.

Résumé étendu. – Des travaux récents, basés sur des données de terrain et de subsurface en Sicile et dans l'Apennin du Sud, montrent que durant la convergence entre les plaques Afrique et Europe (fig. 1) les sédiments déposés sur la marge africaine et les fonds océaniques de la Néotéthys, actuellement subduits, ont été accrétés contre la marge européenne au Néogène. Cet ensemble constitue aujourd'hui trois prismes d'accrétion-collision juxtaposés latéralement, du nord-est au sud-ouest : l'Apennin, la ride calabraise et la Sicile [Roure *et al.*, 1990].

Des échantillons ont été prélevés au niveau des chevauchements de base de l'Unité tectonique inférieure, au pied du M[t] Scalpello, dans la partie orientale du domaine de Caltanissetta (fig. 1). Les échantillons analysés sont des blocs de radiolarite provenant d'un niveau interstratifié dans une matrice argileuse écailleuse (d'âge crétacé inférieur). Cette formation (mélange varicolore inférieur, fig. 1) est disposée en lentilles de 30 à 50 m au pied de la barre de calcaire triasique du M[t] Scalpello et souligne la zone de chevauchement [Larroque, 93].

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Les blocs sont constitués par des éléments de radiolarites, dispersés au sein de veines de quartz et de calcite (pl. I : 1). La largeur des veines est comprise entre 1 et 20 mm et le volume moyen occupé par ces minéralisations varie de 20 à 40 % suivant les échantillons. Les veines sont plus ou moins déformées (veines plissées et cisaillées, déformation des macles de la calcite, décalage des différentes générations de veines). Certains blocs montrent aussi des injections d'argile entre les veines et les éléments de radiolarites (pl. I : 3). Toutes ces figures, typiques de l'éclatement des blocs, nous amènent à les interpréter comme des brèches résultant de la fracturation hydraulique du niveau de radiolarites initial. Quelques échantillons permettent de reconnaître une chronologie parmi les différentes générations de veines. Le quartz est le premier à cristalliser, il est suivi par au moins deux générations de calcite (calcite I et II). La première emprunte partiellement le réseau des fentes de quartz alors que la seconde emprunte le réseau de la calcite I ou recoupe l'ensemble.

Une étude détaillée de l'ensemble de ce secteur montre que les minéralisations sont localisées uniquement au niveau des chevauchements. Nous les considérons donc comme des veines syntectoniques contemporaines du fonctionnement de ces zones de faille [Roure *et al.*, 1990; Larroque, 1993].

Dans les veines de quartz et de calcite échantillonnées dans le chevauchement basal, les fluides associés à la minéralisation ont été piégés en différentes générations d'inclusions fluides. Les inclusions aqueuses primaires contiennent des solutions de faibles salinités (proches de 0,8 équiv. wt % NaCl) dont les températures d'homogénéisation, mesurées par microthermométrie, présentent des valeurs croissantes de 90°C dans le quartz aux épontes à 180°C dans la calcite centrale. Dans les veines de calcite I et II, des inclusions contenant des hydrocarbures légers en C8-C12 ont été caractérisées par microspectrométrie infra-rouge (FTIR) et microfluorescence UV. Elles attestent de la présence dans le fluide minéralisateur, de matière organique ayant dépassé le stade de maturation de la fenêtre à huile.

Le calcul des conditions pression-température, à partir de l'évolution des isochores des inclusions aqueuses, compte tenu de la pression minimale (pression lithostatique + poids de la colonne d'eau au moment de la déformation) de 0,8 à 1 kbar nécessaire à la formation des brèches hydrauliques, conduit à des valeurs de piègeage de 140°C dans le quartz à 235°C dans la calcite. Le recoupement de l'isochore calculé d'une huile légère à faible teneur en méthane, proche de celle contenue dans les inclusions d'hydrocarbures avec celui des solutions aqueuses amène à des valeurs comparables. De plus, des particules de pyrobitumes, associées aux veines de calcite, indiquent qu'elles ont atteint une température d'au moins 150°C. On ne les retrouve pas dans les veines échantillonnées dans les zones de chevauchement plus superficielles du mélange varicolore supérieur, où les inclusions aqueuses sont essentiellement monophasées et de faible salinité. Elles témoignent de circulations de fluides de basse température d'environ 50-60°C. On n'y observe pas d'inclusions d'hydrocarbures.

Le spectre X des argiles du mélange encaissant des brèches hydrauliques présente des indices de transformation thermique des minéraux argileux (I/S de type R_3) situés uniquement au contact de la brèche alors que l'ensemble du mélange est constitué de minéraux argileux I/S de type R_0.

La stricte localisation des veines au niveau des zones de failles suggère que l'expulsion des fluides, lors de la formation du prisme sicilien, s'est effectuée le long des zones de forte perméabilité (chevauchements et décollements). Des fluides chauds et très peu salés ont circulé dans la zone de chevauchement basal, connectée au décollement inférieur du prisme, et provoqué la formation des brèches hydrauliques. Le déséquilibre thermique entre l'encaissant des brèches et les fluides piégés suggère que ces fluides n'ont pas été chauffés *in situ*, mais ont migré le long du décollement, depuis des zones plus internes et plus profondes du prisme.

Ces résultats nous amènent à poser un certain nombre de problèmes concernant : (1) la raison de l'absence de veines minéralisées hors des zones de faille ; (2) la source de chaleur et l'origine des fluides dilués ainsi que ; (3) l'origine des hydrocarbures (roche mère potentielle) et leur mécanisme de migration. Ils seront abordés dans une étude plus générale en relation avec un modèle de circulation de fluide proposé pour le front du prisme sicilien (Larroque *et al.*, en préparation).

I. – INTRODUCTION

Subduction wedges are built up by accretion and deformation of water-rich sediments. Numerous data show that sediments entering subduction zones contain, on average, more than 50 % water and lose most of their fluids during subsequent deformation [Bray and Karig, 1985]. For wedges containing shaly layers, the dewatering seems to be controlled by faults zones [Le Pichon *et al.*, 1986; Moore, 1989; Le Pichon *et al.*, 1993]. In this work, we aim to assess the importance of the fluid distribution and dynamics in the deformation processes of the Sicilian wedge.

The Sicilian wedge (fig. 1) was built up in a mixed subduction and collision context where the tectonic processes are similar to those occurring within modern oceanic accretionary prisms [Roure *et al.*, 1990]. Outcrops of the décollement zones are exposed, in the frontal part of the wedge, allowing direct structural studies and sampling. We focused our sampling on the syntectonic veins and on the surrounding shales of the décollement in order to study the fluid inclusions and determine the physical and chemical properties of the paleo-fluids (composition, P/T conditions of trapping).

Since thermodynamic data are available for many types of fluid systems, it is possible to carry out a rigorous treatment of the fluid inclusion data [Roedder, 1984]. These results are important for constraining the thermal modelling of the shallow part of subduction zones. The present data relate to a Plio-Pleistocene décollement zone, and thus are useful for comparison with modern accretionary wedges such as Barbados [Foucher *et al.*, 1990] or Nankai prism [Yamano *et al.*, 1992].

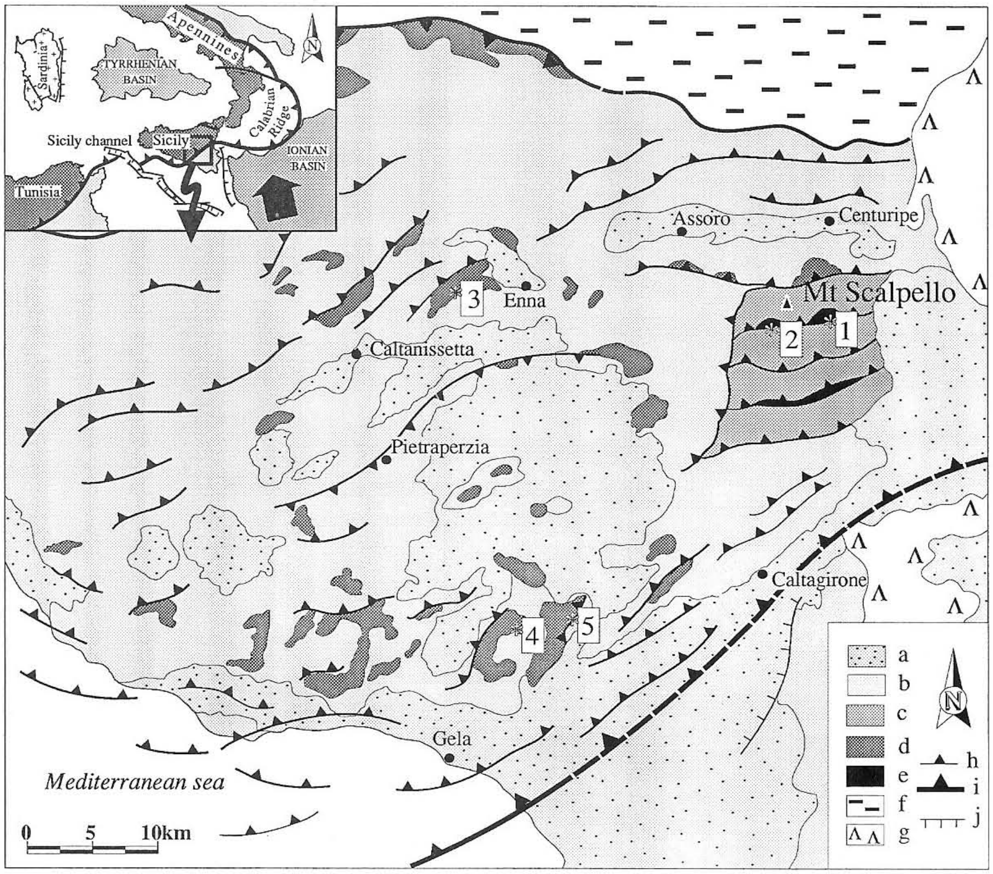


FIG. 1. – Structural map of central Sicily. a : piggyback basins (late Pliocene-early Pleistocene) ; b : Upper Tectonic Unit (late Oligocene to early Pleistocene) ; c : Lower Tectonic Unit (Trias to middle Miocene Mt Scalpello duplex zone) ; d : upper varicoloured mélange (late Cretaceous-middle Eocene) ; e : lower varicoloured mélange (early Cretaceous) ; f : internal unit (Panormide) ; g : Plio-Quaternary volcanism ; h : thrust ; i : deformation front ; j : normal fault. 1, 2, ..., 5 : sampling sites.

FIG. 1. – *Schéma structural de la Sicile centrale.* a : *bassins flottants (Pliocène supérieur-Pléistocène inférieur) ;* b : *unité tectonique supérieure ;* c : *unité tectonique inférieure ;* d : *mélange varicolore supérieur (Crétacé supérieur-Eocène moyen) ;* e : *mélange varicolore inférieur (Crétacé inférieur) ;* f : *unité interne (Panormide) ;* g : *volcanisme plio-quaternaire ;* h : *chevauchements ;* i : *front de déformation ;* j : *faille normale.* 1, 2, ..., 5 : *sites d'échantillonage.*

II. – TECTONIC SETTING

The Apennine-Sicilian belt is part of the complex Alpine collision boundary (fig. 1) between the African and European continents [Roure *et al.*, 1990; Casero *et al.*, 1988]. It is composed of the sedimentary cover of the southern margin of Tethys, the ocean basin between Africa and Europe which occurred in the Jurassic and was later subducted [Dercourt *et al.*, 1986]. The upper plate, occupying most of Sicily, is made of a post-Miocene accretionary wedge. The tectonic imbrications (offscraping and underplating) began during the early Miocene, continuing at least through the early Pleistocene and probably up to the present [McKenzie, 1972]. The southern tip of Sicily corresponds to the poorly deformed African plate (Ragusa foreland). The present study was undertaken in the central Sicilian domain, the most frontal unit of the belt. This domain (fig. 1 and 2) displays Plio-Quaternary thin-skinned tectonics, which affect Triassic limestones and Cretaceous to Quaternary turbidite-rich sequences [Lentini *et al.*, 1974 ; Di Geronimo *et al.*, 1978 ; Bianchi *et al.*, 1987]. The imbricate stack comprises a lower and an upper thrust-sheet units (fig. 2). The lower thrust-sheets (Middle Triassic to Middle Miocene material) make up tectonic duplexes [Bianchi *et al.*, 1987 ; Roure *et al.*, 1990]. At the base of these duplexes (M[t] Scalpello area, fig. 1) lenses of varicoloured mélange (lower mélange), which are involved in the basal thrust fault zone, crop out (fig. 1).

The upper thrust-sheets (late Oligocene to early Pleistocene material) generally lie on top of a décollement zone which contains a varicoloured mélange [upper mélange, Roure *et al.*, 1990 ; Larroque, 1993] composed of red and green clayey matrix with a typical "blocks in matrix" fabric [Cowan, 1985].

The biostratigraphic age of the varicoloured mélange ranges from early Cretaceous (lower varicoloured melange) to Upper Cretaceous-Middle Eocene (upper varicoloured melange), for both the matrix and the blocks [Larroque, 1993]. Blocks are marine sandstones, mudstones, limestones and radiolarian cherts. The matrix is intensively sheared and microfolded, displaying the typical structure of brittle-ductile shear zones [Prior and Berhmann, 1990]. Many of the blocks are deformed, showing veins of quartz and/or calcite.

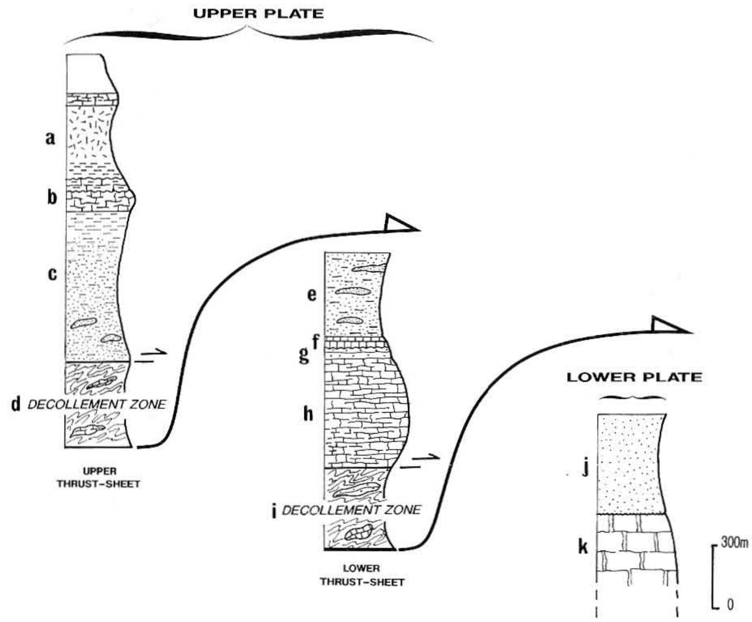


FIG. 2. – Simplified tectonostratigraphic successions. a : late Pliocene to Quaternary shales and sands ; b : Messinian to early Pliocene limestone and evaporites ; c : late Oligocene to late Miocene turbidites ; d ; late Cretaceous to middle Eocene varicoloured mélange ; e : late Oligocene to middle Miocene turbidite ; f : Eocene pelagic limestones ; g : Jurassic radiolarian cherts ; h : Triassic limestones ; i : early Cretaceous varicoloured mélange ; j : Plio-Quaternary sand and shales ; k : Miocene limestones.

FIG. 2. – *Colonnes tectonostratigraphiques simplifiées.* a : *Pliocène supérieur à Quaternaire (argiles et sables) ;* b : *Messinien à Pliocène inférieur (calcaires et évaporites) ;* c : *Oligocène supérieur à Miocène supérieur (série turbiditique) ;* d : *Crétacé à Eocène moyen (mélange varicolore) ;* e : *Oligocène supérieur à Miocène inférieur (série turbiditique) ;* f : *calcaire pélagique éocene ;* g : *Jurassique (radiolarites) ;* h : *Trias (calcaires) ;* i : *Cretacé inférieur (mélange varicolore) ;* j : *Plio-Quaternaire (sable et argiles) ;* k : *Miocène (calcaire de plate-forme).*

Furthermore, in the most frontal part of the belt, shale diapirism of the mélange suggests the presence of overpressured fluids in the vicinity of the décollement [Larroque *et al.*, 1991].

III. – SAMPLING AND RELATIONSHIP OF VEINS TO DEFORMATION

Mineralized veins occur mainly within the varicoloured mélange, at the bases of the lower and the upper thrust-sheets, large sized veins (1 centimetre size) are localized in the blocks of the mélange, smaller ones (1 millimetre size) are interbedded in the scaly planes of the matrix. Few are in continuity from the blocks to the matrix (10 to 20 centimetres length). No significant mineralizations are found in the thrust-sheet units.

— In the lower mélange, 10 samples were collected in the thrust fault zone at the base of the lower unit (south of the M[t] Scalpello, sites 1 and 2, fig. 1). A complex chert level (30 to 40 cm thick) is embedded in the scaly clay matrix. Brecciated beds display a typical "puzzle" structure with clasts of chert and extensional veins filled by mineralizations (pl. I : 1, 2 and 3). The clasts are mostly angular

PL. I. – PLATE I

1. – Hydraulic breccia of the lower mélange showing different generations of veins : quartz calcite I and calcite II (site n° 1).

1. – *Brèche hydraulique du mélange inférieur présentant les différentes générations de veines : quartz, calcite I et calcite II (site n° 1).*

2. – Thin-section microphotography showing quartz veins, calcite I and calcite II veins in hydraulic breccia (site n° 1).

2. – *Photographie (lame mince) de la géométrie des veines de quartz, des veines de calcite I et II dans les brèches hydrauliques (site n° 1).*

3. – Hydraulic breccias of the lower mélange (chert clasts) : a : chert ; b : quartz ; c : clays flowage into the breccia (site n° 1).

3. – *Brèche hydraulique du mélange inférieur :* a : *morceau de radiolarite ;* b : *quartz ;* c : *argile injectée dans la brèche (site n° 1).*

4. – Extrados tension veins with calcite mineralization in a folded blocks of the upper mélange (site n° 5).

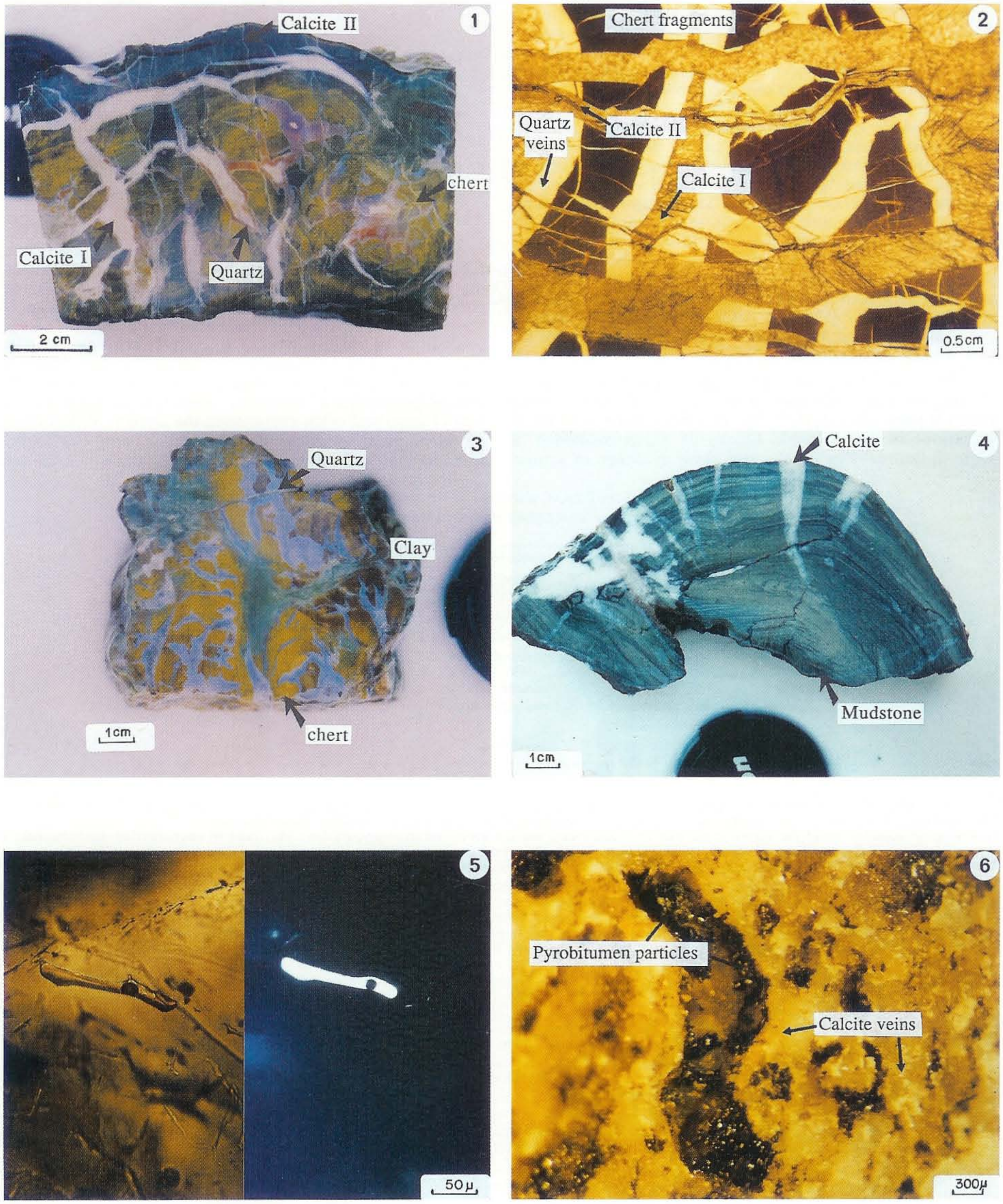
4. – *Veine de calcite sur l'extrados d'un pli dans un bloc du mélange supérieur (site n° 5).*

5. – Thin-section microphotography of hydrocarbon fluid inclusion in sparry calcite vein : in transmitted light (left) and UV light (right) (site n° 1).

5. – *Microphotographie (lame mince) d'une inclusion fluide à hydrocarbure dans une veine de calcite sparitique : en lumière transmise (à gauche), en éclairage UV (à droite) (site n° 1).*

6. – Pyrobitumen particle in a calcite vein (site n° 1).

6. – *Particule de pyrobitume dans une veine de calcite (site n° 1).*


1
Calcite II
chert
Calcite I
Quartz
2 cm
2
Chert fragments
Quartz veins
Calcite II
Calcite I
0.5 cm
3
Quartz
Clay
chert
1cm
4
Calcite
Mudstone
1cm
5
50µ
6
Pyrobitumen particles
Calcite veins
300µ

and do not show any significant displacement, rotation or internal deformation. The size of the clasts range from a few millimetres to few centimetres.

Photo 1 illustrates the relative chronology of veining in the radiolarian chert boudins. Quartz is first to crystallize around the patches of radiolarian chert in a vuggy network. The geometry of this network appears complex with veins parallel, perpendicular and oblique to the bedding and suggest some bursting of the rock. Next, an early generation of large calcite veins partly intrudes and crosscuts the previous quartz veins. Many of the calcite twins are strained. This pattern is crosscut, perpendicular to the bedding, by at least one further generation of calcite veins (photo 2). Numerous features of twinning or faulting and folding veins are major criteria to assume that these veins are syntectonic. Some samples show mud injection through the clasts and the veins (photo 3). The transition to the bedrock is progressive, but some veins penetrate a few centimeter into the scaly clay bedrock. Since these features are typical of hydraulic breccias [Phillips, 1972; Sibson, 1986; Jebrak, 1992], we infer that fluids cause hydraulic fracturing of the rock during deformation.

— At the base of the upper thrust-sheet units, 12 samples were collected in the upper mélange at three different sites (sites 3, 4, 5, fig. 1). Only calcite-type mineralizations is found inside blocks of green mudstone or white limestone. The blocks also show evidence of syntectonic crystallization [extrados tension veins (photo 4), faulted and folded veins], but no features typical of hydraulic fracturing are present.

IV. – Thermal evolution of the rocks

A) Petrography of the veins

Petrographic observations associated with fluid inclusions studies were carried out on 2 sets of samples (table I).

In the hydraulic breccias of ***the lower mélange***, a fibrous red or grey chalcedony crystallize early as tiny rims around the brecciated fragments. Then microcrystalline quartz occurs in veins with increasing size in the centre. X-ray analysis does not indicate the former presence of opal. The first generation of calcite crystals (calcite I) are pure sparitic-type in large veins (around 10 mm or more) often occurring through the path positionned first by quartz veins. The second generation of carbonate veins (calcite II) is smaller size (1 mm) and often crosscuting the former. SEM and microprobe analyses show varying Mn content (up to 5 % MnO in the calcite II). The width of the veins is 1-20 mm.

TABLE I. – Type of inclusions in the studied samples.

TABLE I. – Type d'inclusions dans les roches échantillonnées.

	Aqueous singlephase inclusions	Aqueous twophase inclusions	Hydrocarbon inclusions	Bitumen particles
Lower mélange (sites 1, 2) Samples N°	/	CalciteI and II and quartz veins (chert clasts) 25-196, 27-196, 12-93, 14-93, 16-93	Calcite I and II veins (chert clasts) 25-196, 27-196, 37-198	Calcite(I) and II veins (chert clasts) 8-93, 13-111
Upper mélange (sites 3, 4, 5) Samples N°	Calcite veins (Cretaceous green mudstone) (Eocene white limestone) 72-177, 73-177, 10-1, E6-1, E4-2, E6-2, E3-57, E4-12	Calcite veins (Cretaceous green mustone) 9-1, E4-4	/	/

TABLE II. – Estimated mineralogical composition of bulk rock based on intensities ratio of selected peaks to the 4.27 Å peak of quartz : IM : illite/mica; K/C : kaolinite; C : chlorite; Qz : quartz; Cc : calcite; Dol : dolomite; Hmt : hematite; F : feldspar; Goet, goethite.
Mineralogical composition of the clay-size fraction (< 2 µm), from ethylen-glycol solvated preparation : IS_0 : random interstratified illite/smectite; I : expandable illite; % Sm : percent smectite layers determined from angular position of the $\theta(003)_{17}$ peak and the $\theta(001)_{10}$ peaks; R_0, random organisation; R_3, ordered organisation of the IMII type. K/C, undifferentiated Kaolinite/Chlorite; M, 10 Å phase.

TABLE II. – *Composition minéralogique approchée de la roche totale (basée sur le rapport des intensités du pic sélectionné avec le pic à 4,27 Å du quartz.) : I/M : illite/mica ; K/C : kaolinite ; C : chlorite ; Qz : quartz ; Cc : calcite ; Dol : dolomite ; Hmt : hématite ; F : feldspath ; Goet : goethite. Composition minéralogique de la fraction argileuse (< 2 µm), après traitement à l'éthylène-glycol :* IS_0 : *interstratifié désordonné illite/smectite ; I. illite gonflable ;* % Sm : *pourcentage de smectite déterminé à partir de la position angulaire du pic* $(003)_{17}$ *pour l'interstratifié et* $\theta(001)_{10}$ *pour l'illite gonflante ;* R_0 : *organisation désordonnée ;* R_3 : *organisation ordonnée de type IMII.* K/C : *kaolinite/chlorite indifférenciée ;* M : *phase 10 Å.*

Mineralogical composition		**Clay-size fraction < 2µm**					**Bulk Rock**								
SAMPLE N°	OUTCROP N°	IS_0 $\theta(003)_{17}$	%Sm	I $\theta(001)_{10}$ %Sm	K/C I_7/I_{17}	M I_{10}/I_{17}	Clay 12-15Å	IM 10Å	K/C 7Å	Qz 4.27Å	Cc 3.02Å	Dol 2.89Å	Hmt 2.70Å	Goet 4.18Å	F 3.12Å
LOWER MELANGE															
32-196	1	R_0	10	10	10					1			0.71	2	
5-93	1	R_0	44	14	10					1	2.25		0.60		
10-93	1	R_3		10			0.56	0.27	0.67	1	0.33	1.33	0.40		0.44
UPPER MELANGE															
7-1	3	R_0	72		16	17	0.55	0.23	0.45	1	4.95				
21-193	4	R_0	55		56	31	0.37	0.19	0.85	1	0.04				0.22
13-93	4	R_0	40		33	31	0.50	0.29	0.89	1	0.33		0.25		0.36
44-65	4	R_0	68		76	38	0.25	0.21	0.70	1	1.17		0.18		0.26
7-191	5	R_0	80		11	14	1.33	0.67	0.83	1	2.27	0.83			0.83
8-191	5	R_0	68		26	22	0.89		0.68	1			0.40		

while the mean volume occupied by the mineralizations varies between 20 and 40 %.

In *the upper mélange*, the mineralizations are uniformly distributed in all the blocks, making up 10 % by volume of the rocks. The crystals are pure calcite sparry-type. The width of the veins is 1 to 20 mm.

B) The shaly bedrock

Samples of clayey mélange matrix were analysed by X-ray diffraction for each site [Nicot *et al.*, 1992].

In the lower mélange : clay mélange matrix samples were collected at the contact with the hydraulic breccia (10-93, table II) and some tens of centimetres from the veins within the breccia (5-93, 32-196). The bulk mineralogy contains abundant calcite and minor felspar. Chlorite and kaolinite are either distinguished by two peaks at 3.5 Å, or considered as an undifferentiated K/C phase.

The less than 2 µm fractions are quite different : a random interstratified illite/smectite occurs in the samples (5-93, 32-196) within the clasts, whereas an ordered interstratified illite/smectite of R_3 type is present in contact with the veins in sample 10-93.

In the upper mélange (7-1, 7-191, 8-191, 21-193, 13-93, 44-65, table II) samples were also collected adjacent and farther away from the veins.The main difference in the bulk rock mineralogy is that the matrix is mostly devoid of calcite.

The fraction less than 2 µm contains quartz in addition of the clay minerals. The dominant clay mineral is a random interstratified illite/smectite, with a broad range of smectite layers (80 to 40 %). Kaolinite is common. Heating treatments and observation of the (060) peak position show that chlorite is absent or undetectable in this fraction.

V. – Results of palaeo-fluid studies

As fluid inclusions result from the trapping of fluids during ancient fluid circulation, the characteristics of the inclusions most commonly entrapped in syntectonic mineralizations can provide direct informations on the nature and P/T conditions of the fluids circulating during deformation [Roedder, 1984]. On the other hand, the characteristics of organic matter associated within mineralizations can give additional information on the temperature. In this study, aqueous syngenetic fluid inclusions were commonly found in quartz and calcite, and hydrocarbon fluid inclusions (pl. I : 5) associated with bitumen particles (pl. I : 6) were observed in the calcite veins of chert clasts (lower mélange, table I).

Using different analytical techniques, an attempt was made to define the composition of the fluids and their conditions of flow in the décollement zones during deformation [Larroque *et al.*, 1992 ; 1993]. To be consistent with the constant volume assumption, investigations were conducted on sheltered zones where microscopic observation does not show petrographic indications of stretching or leakage.The details of the analytical techniques are given in the appendix.

A) Fluid inclusion studies

In the lower mélange, fluid inclusions in quartz and calcite are most frequently found in hydraulic breccias within chert clasts.

— *Quartz veins* (25-196, 27-196, 12-93, 14-93, 16-93, table I), contain aqueous primary fluid inclusions in growth zones and as isolated cavities. They are of small size (5 to 10 µm) and oftenly clustered in the inner part of the crystals and in growth zones in the external part. The inclusions are biphase and show relatively homogeneous filling ratios. The homogenisation temperatures (T_h) obtained for these inclusions by microthermometry [Guilhaumou, 1982] are less than 100°C, with a mode between 70 and 90°C (fig. 3A). The inclusions trapped near the rims of quartz crystals have the highest T_h values (near 90°C). Microcryoscopic analyses (fig. 3B) yield very homogeneous last melting temperature of ice (T_m with an average (around – 1°C), which is in favor of a well preserved content. This corresponds to a low salinity water with a composition in the

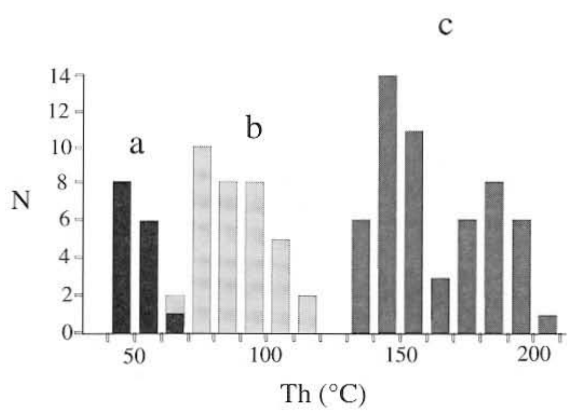


Fig. 3A

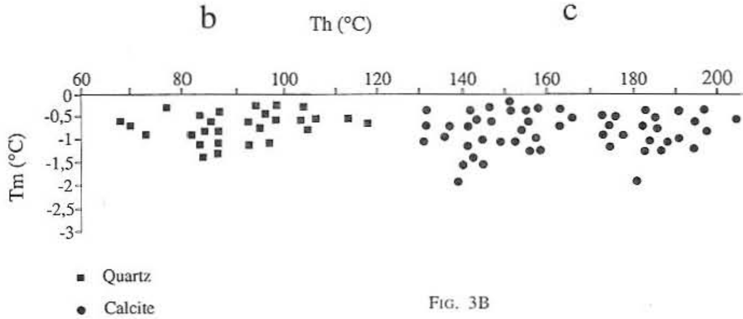


Fig. 3B

Fig. 3. – A. – Homogenisation temperatures. a : hydrocarbon-bearing fluid inclusions in calcite ; b and c : aqueous fluid inclusions in quartz and calcite veins (hydraulic breccia).
B. – T_m/T_h diagram for fluid inclusions in quartz and calcite veins (hydraulic breccia).

Fig. 3. – A. – *Températures d'homogénéisation.* a : *inclusion d'hydrocarbure dans la calcite ;* b *et* c : *inclusions fluides aqueuses dans les veines de quartz et de calcite (brèche hydraulique).*
B. – *Diagramme de corrélation T_m/T_h pour les inclusions fluides dans les veines de quartz et de calcite des brèches hydrauliques.*

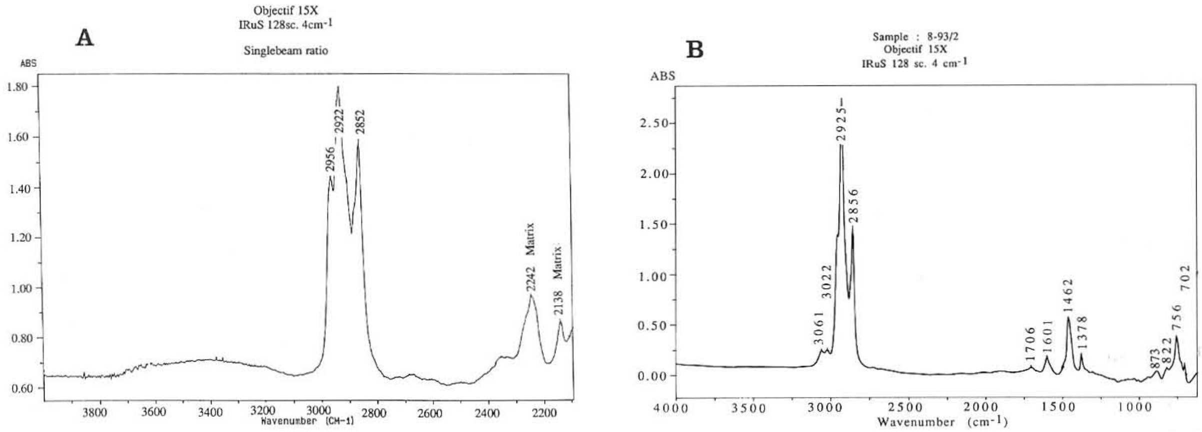


FIG. 4. – A. – MicroFTIR spectra of hydrocarbon fluid inclusions. The spectrum shows characteristic vibrations of aliphatic components : 2852-2922-2956 cm^{-1} (C-H aliph) and of the matrix 2138-2242 cm^{-1} (quartz).
B. – MicroFTIR analysis on bitumen particles. The spectrum show characteristic vibrations of aromatic components at 760-820-870 cm^{-1} (aromatic C-H deformation bands), 1601 cm^{-1} (C = C streching of the rings), 1706 cm^{-1} (C = O stretching) and 3020-3060 cm^{-1} (C-H stretching). The other bands between 2800-3000 cm^{-1} and 1378-1463 cm^{-1} correspond to stretching and bending of aliphatic C-H respectively.

FIG. 4. – A. – *Spectre microFTIR pour les inclusions fluides à hydrocarbure : le spectre montre les vibrations caractéristiques des composés aliphatiques : 2 852-2 922-2 956 cm^{-1} (C-H aliph) et de la matrice 2 138-2 242 cm^{-1} (quartz).*
B. – *Spectre microFTIR pour les particules bitumineuses : Le spectre montre les vibrations caractéristiques des composés aromatiques : 760-820-870 cm^{-1} (déformation des C-H), 1 601 cm^{-1} (élongation des C = C), 1 706 cm^{-1} (élongation des C = O) and 3 020-3 060 cm^{-1} (élongation des C-H). Les autres bandes entre 2 800-3 000 cm^{-1} and 1 378-1 463 cm^{-1} correspondent respectivement aux élongations et aux déformations des C-H aliphatiques.*

H_20-NaCl-(KCL) system of 1.651 equiv.wt % NaCl and a density of 0.972 g/cm^3 [Brown and Lamb, 1989]).

— *Aqueous large primary intracrystalline inclusions*, about 15-20 µm in size, are more uniformly distributed in calcite crystals (25-196, 27-196, 12-93, 14-93, 16-93, table I). They are biphase, irregularly shaped and often representative of dendritic growth. The liquid/vapour ratios are relatively homogeneous for well preserved inclusions. The Th values range from 120 to 200°C, with two modes, near 140 and 180°C (fig. 3A). The Tm values, are higher than – 2°C, also indicating a low salinity water (fig. 3B). The composition of the fluid also lies in the H_2O-NaCl-KCl, system with 0.827-1.651 equiv. wt % NaCl and corresponding densities of 0.94-0.89 g/cm^3.

SEM-EDS analysis on frozen inclusions [Ayora and Fontarneau, 1990], was used to compare the spectra from dilute sea water (standard solution with 1/2, 1/4, 1/8 and 1/16 dilution) and the spectra obtained from inclusions in quartz and calcite veins. The results show that the salinity in these inclusions is lower than 8 g/l. This is very close to the value of equiv. 0.827 equiv. wt % NaCl calculated from microcryoscopic data.

CH_4, N_2, H_2S, SO_2, CO_2 and light hydrocarbons were not detected by microRaman spectrometry (following conditions defined by Burke and Lustenhower [1987]) in the gaseous phases or as dissolved components in the aqueous phases of fluid inclusions in any sample. For CH_4, the detection limit, in the operating condition, was near 0.04 mole %.

— *In the calcite veins* (25-196, 27-196, 37-198, table I), and at the quartz/calcite interfaces, large hydrocarbon inclusions (20-30 µm) displaying UV fluorescent dull blue colour in are observed (pl. I : 5). These inclusions are partly single-phase. When they are biphase, the T_h are ranging from 50 to 60°C (fig. 3A). These hydrocarbons were analysed by micro FTIR. The type of spectra registered (fig. 4A) show the characteristic stretching vibrations of aliphatics between 2 800 and 3 000 cm^{-1} [Colthup *et al.*, 1990]. CH_4, CO_2, aromatics, water and sulphur components were not detected. This spectra is representative of a light aliphatic oil depleted in CH_4 and CO_2. The equivalent n-alkanes calculated are in the range C_8-C_{12} [Guilhaumou *et al.*, 1990]. These hydrocarbon bearing fluid inclusions are mostly abundant in calcite II. Their random distribution (pl. I : 5) allow to presume they are contemporaneous of aqueous primary inclusions.

In the upper mélange, primary aqueous fluid inclusions in calcite veins (72-177, 73-177, 10-1, E6-1, E6-2, E4-2, E3-57, E4-12, table I) are mostly liquid single-phase at room temperature. The few measurements obtained in seldom biphase inclusions (Cretaceous green mudstone blocks; 9-1, E4-4, table I) show homogenisation temperatures near 50-60°C.

Some large and flat-shaped aqueous single-phase inclusions were observed in numerous calcite samples of all blocks. These inclusions forming healed shear planes are regularly spaced within the planes from cleavages steps and thus considered as secondary and characteristic of very late-stage fluid during brittle deformation [Roedder 1984].

B) Organic matter studies

In the lower mélange, the calcite veins contain numerous solid black particles (8-93, 13-111, table I; pl. I : 6), that were easy to isolate from the rock for direct analysis [Guilhaumou and Larroque, 1993]. Rock Eval pyrolysis, applied on isolated particles from two different samples (table III), give an HI averaging 578. This is a typical value for organic matter of marine origin. The average t°max values at 445 ± 1°C indicate hydrocarbons in the oil window [Espitalié *et al.*, 1985; Espitalié *et al.*, 1986]. MicroFTIR analyses were obtained on very thin particles by crushing them in a diamond cell to avoid detector saturation. The spectra

TABLE III. – Rock Eval analyses of bitumen particles.
TABLE III. – *Analyses Rock Eval des particules bitumineuses.*

Calcite I -II Sample N°	weight for process	S1	S2	S3	TOC	t°max	HI	OI
8-93	11,1mg	38,10	277,20	2,61	48,24%	447°C	574	5
13-111	10,6mg	48,11	360,00	2,64	61,73%	444°C	583	4
Mean		43,1	318,6	2,62	54,98%	445°C	578	4,5
		mgHC/gROCK	mgHC/gROCK	mgCO2/gROCK			mg HC/g TOC	mg CO2/g TOC

obtained (fig. 4B) show aromatic vibrations characteristic of an organic component with a high degree of maturation [i.e. bitumens, Colthup *et al.*, 1990].

VI. – INTERPRETATION AND DISCUSSION

It is now well established in the scientific litterature, that fluid inclusions in quartz veins yield valid information, yet fluid inclusions in calcite crystals remain underutilized and need to be discussed. Numerous analytical and experimental studies have been published on this subject [e.g. Guilhaumou *et al.*, 1984; Gratier and Jenatton, 1984; Prezbindowski and Larese, 1987; Bakker, 1992]. In fact these authors have shown that, in the absence of significant reheating or large changes in pressure (i.e., high pressure metamorphism), the inclusions are able to preserve the physico-chemical parameters (composition and density) of the initial fluids [Lefaucheux *et al.*, 1972]. In the present study, there is no reason from geological history or petrographic observations to suggest any heating above trapping temperature or a large drop in pressure after the inclusions formation. The difference in the T_h values of quartz and calcite (about 60°C) is not sufficient in view of the inclusion size in quartz (5-10μm) and of the minimum confining pressure at time of vein formation (near 0.8-1 kbar), to have induced an equilibration of the inclusions in quartz [Pecher and Bouiller, 1984]. Furthermore, careful microscopic and SEM examination do not show any evidence for stretching or decrepitation in these samples, as observed by Bakker [1992].

Therefore, we propose that the fluid inclusion data obtained from the chert clasts, as indicated previously are indicative of an increase in temperature (from quartz to calcite) during veining.

A) P/T conditions : fluid inclusions and organic matter data

In samples from the lower mélange, the previously discussed relationships of the veins to deformation show that the fluids responsible for the quartz and calcite veining were entrapped during hydraulic fracturing of the chert. We may assume that the fluid pressure during fracturing was near the lithostatic pressure [Sibson, 1986]). Restoration of the lithostratigraphic column and hydrostatic water column at time of deformation yield a reasonable minimum estimation of depth for the lower fault zone (upper thrust-sheet + lower thrust-sheet) : near 2,500-3,000 m. For rocks at 2,500-3,000 m with a density of 2.4 under 1,000-2,000 m water depth, we then computed a lithostatic pressure value of 0.8-1 kbar. Following the work of Phillips [1972], we admit that when the rock is hydraulically fractured in a normal fault setting there is an abrupt drop in pressure due to opening of the fractures, but in the case of hydraulic fracturing related to thrust fault, however the pressure remain, at least, equal to the lithostatic load.

Using the isochores corresponding to densities of 0.94 and 0.89 g/cm³ in the H_2O-NaCl-KCl system [Brown and Lamb, 1989], for inclusions of salinity 0.827 wt % eq. NaCl, the average trapping temperatures (Tt) can be calculated from these pressure values. They fall in the range of 140-150°C for inclusions in quartz veins and 180-235°C for inclusions in calcite veins (fig. 5). The hydrocarbon-bearing fluid inclusions observed in calcite veins appear often as cogenetic with aqueous ones, an attempt was made to compute the liquid/vapour curve and the isochore of a representative oil using the Peng-Robinson equation [Le Cornec *et al.*, in progress]. It may be assumed that the hydrocarbon contained in the fluid inclusions is a light aliphatic oil lacking of significant dissolved gases (i.e. CH_4 or CO_2) by having the overall composition obtained by microFTIR (alkane equivalent : C_8-C_{12}). From preliminary work, we admit that the gas content is the main parameter influencing the position of these curves in P-T paths. Thus, the aqueous and hydrocarbon isochores, for the inclusions entrapped in the same calcite veins intersect at pressure values near those previously determined from the minimum lithostatic pressure (fig. 5).

From comparison with artificial maturation of organic matter and the corresponding maturation stage in the Paris basin, temperatures of at least 150°C, are also supported by the high maturity level of the pyrobitumens entrapped in calcite veins [Guilhaumou and Larroque, 1993].

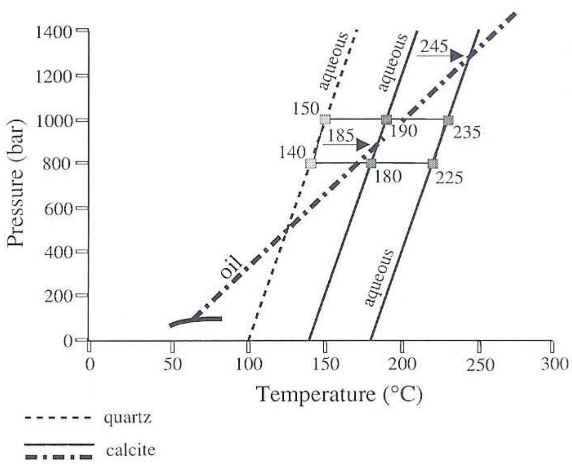


FIG. 5. – Trapping temperature defined from isochores for aqueous fluids in quartz veins ($T_h \cong 100$°C) and in calcite veins ($T_h \cong 140$ and 180°C) for a fluid pressure of 0,8-1 Kb.

FIG. 5. – *Températures de piégeage déduites des isochores des inclusions aqueuses dans les veines de quartz ($T_h \cong 100$°C)* et dans les veines de calcite ($T_h \cong$ *140 et 180°C*) pour une pression de fluide comprise entre 0,8-1 Kb.

In veins from blocks of the upper mélange, mostly single-phase fluid inclusions were found, being interpreted as typical of low temperature fluids. The measurements on seldom biphase inclusions observed indicate temperature near 50-60°C.

B) Incipient and localized transformation of the matrix

Although temperature is not the only parameter controlling the diagenesis of clay minerals [Perry and Hower, 1972; Velde and Vasseur, 1992], it nevertheless remains a major factor. I/S clays of R_0 type in the upper and lower mélange indicate that these shales were not subjected to temperatures above 60-100°C [Velde, 1985].

On the contrary, clays in contact with the hydraulic breccia (I/S of R_3 type) are possibly compatible with the trapping temperatures of fluids in quartz and calcite veins. Thus, if the recrystallization of I/S clays of R_0 and R_3 type is coeval, this increased diagenesis in the shale surrounding could be related to the warm fluids in them. Therefore, a local thermal gradient would exist between the veins and the host-rock. The effect of this heat source would be limited by the initial volume of the warm fluid with the thermal gradient decreasing as a function of time and kinetics.

C) Transient fluid-flow along the basal décollement

The veining occurred during disruption of bedding in the varicoloured mélanges, probably related to the emplacement of the upper and lower thrust-sheets during Plio-Pleistocene imbrications [Roure *et al.*, 1990; Larroque *et al.*, 1991; Larroque, 1993].

The lack of extensive veining inside the thrust-sheet units indicates a negligible diffusive fluid flow throughout the overall wedge. The localization of the veins, found only along the main thrust plane (i.e. varicoloured mélange), shows that the syntectonic dewatering of the Sicilian wedge was essentially controlled by the high permeability décollement zones. Similar observations have been made by Orange *et al.* [1993] in the late Miocene western Olympic accretionary complex, studying the paragenetic sequences of the veins. Furthermore, in the Sicilian case, high temperature freshwater (with a prograde evolution from 150°C in quartz veins to more than 200°C in calcite veins) have been trapped in the lower thrust fault zone connected to the basal décollement [Larroque *et al.*, 1993].

Such a thermal event is not regionally recorded in the surrounding shales, but a local imprint is detected just at the contact of the hydraulic breccia. It may thus be concluded that these fluids were not heated *in situ* but flowed in from a warmer zone, and were possibly channelled along the basal décollement from a deeper internal part of the prism. Thus, the fluid flow was probably rapid and brief, precluding the thermal equilibrium between the shaly matrix and the fluids.

The different nature of hydrothermal minerals (quartz and calcite) and the mutual crosscutting relations between the vein sets (bedding parallel and perpendicular extensional veins, pl. I : 2) suggest multiple episodes of brecciation and hydrothermal recementation. Thus, deformation of the clasts and fluid activity may have continued after the early veining. Such episodic fluid flow along reverse faults has already been proposed by Sibson *et al.* [1988].

CONCLUSION

The chemical composition and the pressure-temperature conditions of fluids active in the Sicilian accretionary prism can be obtained by studying the syngenetic fluid inclusions entrapped within syntectonic veins in the varicoloured melange. The upper mélange contains calcite veins in blocks only, with typical low temperature – low salinity fluid entrapped in fluid inclusions. Temperatures are lower than 100°C and the composition is near fresh water. On the contrary, the lower mélange displays hydraulic breccias with successive episodes of quartz and calcite veining. Two main types of fluid are recognized : (I) a very low salinity water (approximatively 0.8 equiv. wt % NaCl) without significant amounts of dissolved gas (e.g. CO_2-CH_4) in both quartz and calcite and (II) a light aliphatic oil (hosted only in calcite) without significant amounts of gas and relatively depleted in water. Microthermometric measurements indicate high-temperatures low salinity fluids with a prograde evolution from 150°C in quartz, at the onsets of vein formation to more than 200°C in later stage calcite. The calcite veins also display pyrobitumens with a maturation level indicative of high temperature.

The relationships between veining and deformation suggest that the inclusions were entrapped successively during the main deformation episodes. Restoration of the minimal lithostratigraphic succession at the time of deformation yield a minimum associated pressure of 0.8-1 kbar. The pressure-temperature values obtained by intersecting the isochores of aqueous and oil inclusions in calcite minerals fall in the same range. The surrounding shales of the décollement do not record such high temperature but the transformation of clays, near the contact of the veins in the hydraulic breccias, indicates a local thermal imprint.

On the basis of the present data, five main points are established : (1) the localization of syntectonic veins along the décollement zones, (2) multiple episodes of veining in the hydraulic breccias accompanied by an increase in temperature, (3) a difference in temperature between the fluids entrapped in the upper and lower mélanges, (4) a thermal desequilibrium between the host-rock and the fluids in the lower mélange, (5) the presence of oil and pyrobitumens associated with warm fluid in the lower mélange.

Finally, all the data suggest a syntectonic dewatering of the sedimentary succession along the décollement and thrust faults involving a warm transient flow of deep fresh water, channelled along the basal décollement. Therefore, precipitating veins could develop in a fluid flow regime. Either the precipitation occurred during the flow or during the quiet period of this transient regime. In any cases, veining is not the only criteria to suspect fluid flow but the chemical and thermal desequilibrium with the surrounding matrix also support this conclusion. Numerous problems remain, however, concerning the nature of the heat source, the origin of the low salinity water and the organic matter as well as the type of the hydrocarbon maturation process involved (source rocks). These topics are discussed in a more general study which proposes a fluid flow model for the front of the Sicilian prism during Plio-Pleistocene [Larroque *et al.*, in progress].

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