

Diagenetic evolution of fault zones in Urgonian microporous carbonates, impact on reservoir properties (Provence – SE France).

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Microporous carbonate rocks form important reservoir with high a permeability variability depending on sedimentary, structural and diagenetic factors. Carbonates are very sensitive to fluid-rock interactions that lead to secondary processes like cementation and dissolution that modify the reservoir properties. Focussing on fault-related diagenesis, the aim of this study is to identify the fault zone impact on reservoir properties. It focuses on 2 fault zones east to La Fare Anticlinal (SE France) which cross-cut Urgonian microporous carbonates. 122 collected samples along four transects orthogonal to the fault zones were analysed. Porosity values have been measured on 92 dry plugs. Diagenetic elements were determined on 92 thin sections using Polarized Light Microscopy, cathodoluminescence, red alizarin, SEM and isotopic measurements ($\delta^{13}\text{C}$ and $\delta^{18}\text{O}$). Eight different calcite cementation stages and 2 micrite microfabrics were identified. As a main result, this study highlight that the 2 fault zones acted as drain canalizing low temperature fluids at their onset, and induced fault zone calcite cementation during 2 subsequent phases which strongly altered and modified the local reservoir properties.

1. INTRODUCTION

Microporous carbonates form important reservoirs (Deville de Periere et al., 2017; Lambert et al., 2006; Sallier, 2005; Volery et al., 2009), with porosities up to 35% (Deville de Periere et al., 2011). Due to their heterogeneous properties which depend on sedimentary, structural and diagenetic factors, they determine a high variability of reservoir permeability (Bruna et al., 2015; Deville de Periere et al., 2011, 2017; Eltom et al., 2018; Florida et al., 2009; Hollis et al., 2010). Fault zones in carbonates play an important role on reservoir properties (Agosta et al., 2010, 2012; Caine et al., 1996; Delle Piane et al., 2016; Ferraro et al., 2019; Knipe, 1993; Laubach et al., 2010; Rossetti et al., 2011; Sinisi et al., 2016; Solum et al., 2010; Solum and Huisman, 2016; Tondi, 2007; Wu et al., 2019). Fault zones are complex structures composed of damage zones and the fault core encompassed by the host rock (Caine et al., 1996; Chester and Logan, 1986, 1987; Hammond and Evans, 2003). Faults can act as barriers (Agosta et al., 2010; Tondi, 2007), drains (Agosta et al., 2007, 2008, 2012; Delle Piane et al., 2016; Evans et al., 1997; Molli et al., 2010; Reches and Dewers, 2005; Sinisi et al., 2016; Solum and Huisman, 2016), or mixed zones (Matonti et al., 2012) depending of their architecture and diagenetic evolution. Because of their hydraulic properties, fault zones influence the fluid flows in the upper part of Earth crust (Bense et al., 2013; Evans et al., 1997; Knipe, 1993; Sibson, 1994; Zhang et al., 2008), and increase the fluids-rock interactions. Carbonates are very sensitive to these interactions, which lead to secondary processes like cementation and dissolution (Deville de Periere et al., 2017; Fournier and Borgomano, 2009; Lambert et al., 2006). Fault related

diagenesis locally modifies the initial rock properties (mineralogy and porosity), and therefore the reservoir properties (Hodson et al., 2016; Knipe, 1993; Knipe et al., 1998; Laubach et al., 2010; Woodcock et al., 2007). In case of a poly-phasic fault zone, duplication of fluid pathways/barriers lead to very complex diagenetic modifications. The initial vertical and lateral compartmentalization of microporous limestones is, therefore, accentuated by fault-related diagenesis. Hence, understanding of the faulting processes and diagenesis is crucial for a better exploration and production in carbonates. Urgonian microporous carbonates of Provence, are made of facies and reservoir properties analogue to Middle East microporous carbonate reservoirs (Thamama, Kharaib and Shuaiba formations ; Borgomano et al. 2002, 2013; Sallier 2005; Fournier et al. 2011; Leonide et al. 2012; Léonide et al. 2014). Although Urgonian microporous carbonates of Provence are analogue to Middle East reservoirs, the analogy can be extended to other faulted microporous carbonate reservoirs. To have a better comprehension of diagenetic modifications linked to fault zones on these rocks, the aim of this paper is (i) to determine the diagenetic evolution of polyphasic fault zones; (ii) to identify their impact on reservoir properties and (iii) to link the fault evolution with the fluid flow and geodynamic history of the basin.

2. GEOLOGICAL CONTEXT

We studied 2 faults crosscutting microporous Valanginian-to-Early Aptian Urgonian facies carbonates of the South-East basin (Provence-SE France) deposited along the southern margin of the Vocontian Basin (Léonide et al., 2014; Masse and Fenerci Masse, 2011). The “Urgonian” platform carbonates (Masse, 1976) reached their larger extension during the late Hauterivian–Early Aptian (Masse and Fenerci-Masse, 2006). From Albian to Cenomanian, the regional Durancian uplift triggered exhumation of Early Cretaceous carbonates, bauxitic deposits (Guyonnet-Benaize et al., 2010; Lavenu et al., 2013; Léonide et al., 2014; Masse and Philip, 1976; Masse, 1976), and E-W-trending normal faults (Guyonnet-Benaize et al., 2010; Masse and Philip, 1976). During the Late-Cretaceous, platform environment led to a transgressive rudist platform deposition (Philip, 1970). From Late Cretaceous to Eocene, the convergence between Iberia plate and Eurasia plates (e.g. Bestani 2015, and cited references) led to a regional N-S shortening (e.g. Molliex et al. 2011 and cited references). The so-called “Pyrénéo-Provençal” shortening, which gave rise to E-W north-verging thrust faults and ramp folds (e.g. Bestani et al. 2016, and cited references). From Oligocene to Miocene, the area underwent extension associated to Liguro-Provençal Basin opening (e.g. Demory et al. 2011). During Miocene times, the Alpine shortening dimly impacted the studied area (Besson, 2005; Bestani, 2015), and reactivated the “Pyrénéo-Provençal” structures (Champion et al., 2000; Molliex et al., 2011).

We studied 2 faults pertaining to kilometric-scale fault system on the E-W-trending La Fare anticline near Marseille (Fig. 1A). The southern limb of this anticlinal dips 25° S, and is constituted by Upper Hauterivian, Lower Barremian and Santonian rocks (Fig. 1B). The Upper Barremian carbonates are composed, from bottom to top, of a 120m-thick calcarenite unit with cross-beddings, a 40m-thick massive coral-rich calcarenite unit, and a 10m-thick calcarenite unit (Masse, 1976; Matonti et al., 2012; Roche, 2008). Unconformable Santonian rocks are made of coarse rudist limestones (Fig. 1A).

The Castellás fault zone is a kilometre-long strike-slip fault, N060 to 070-trending and 40° to 80°N-dipping (Fig. 2A, 2B; table 1) composed horse structures, secondary faults and lenses (Fig. 2A, 2C; Aubert et al. (2019b)). The second fault zone “D19” is composed of 5 sub-fault zones (F1 to F5) restricted in a 50m-long interval (Fig. 2E, H; Table 1; (Aubert et al., 2019a)). Sub-faults are made of 2 sets. Set one comprises F3 and F4, N040 to N055-trending, 60-80°NW-dipping (orange on Fig. 2F). Set 2 is N030-trending, dipping 80°E, with strike-slip slickensides pitch 20 to 28°SW (F1, F2, F5, red on Fig. 2F). The 5 sub-fault zones show an asymmetric architecture (Aubert et al., 2019a).

The internal structure of both fault zones results from three tectonic events:

- the Durancian uplift dated as mid-Cretaceous leading to extension and to normal *en echelon* normal faults. The Castellás fault is one of them and bear early dip-slip normal striations (Matonti et al., 2012),
- the Early Pyrenean compression with N000° to N170°-trending σ_H (see cited references in Espurt et al. 2012). This event reactivates the Castellás fault as sinistral (Matonti et al., 2012) and leads to the neo-formed strike-slip faults of the D19 outcrop (Aubert et al., 2019a).
- the Pyrenean to Alpine folding, triggering the 25°S tilting of the strata and fault zones. Faults of the D19 outcrop were reactivated while the Castellás fault tilting led to an apparent reverse throw (Aubert et al., 2019a).

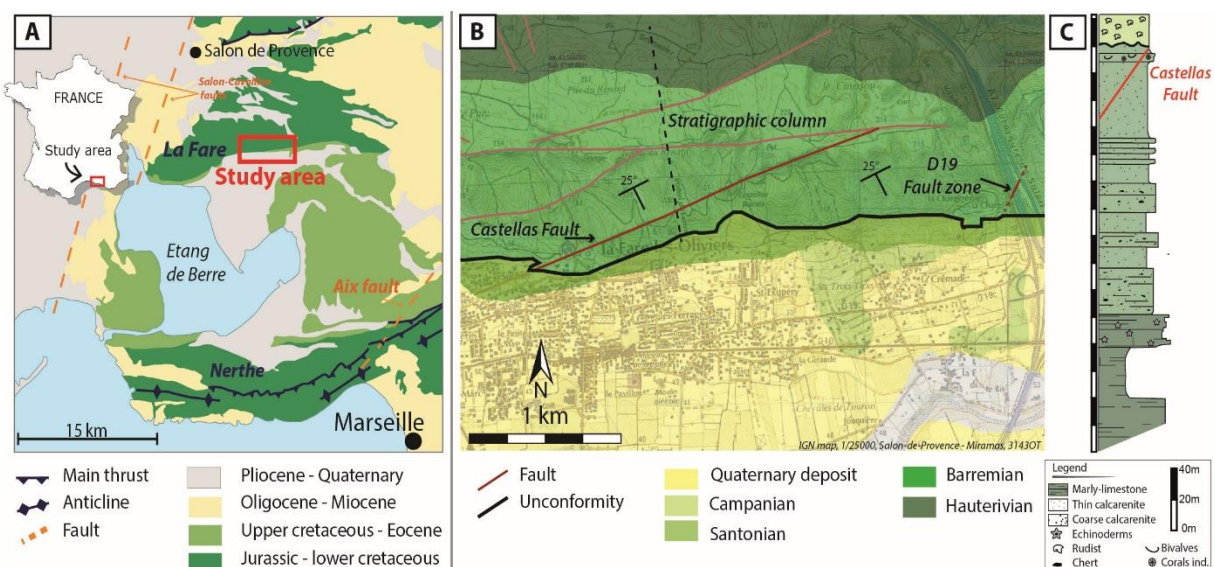


Figure 1 : Geological context of the study area. A: geological map of Provence, B: Simplified structural map with the location of the Castellás fault and the stratigraphic column (black dashed line); B: Stratigraphic column of exposed Cretaceous carbonates (modified from Roche, 2008)

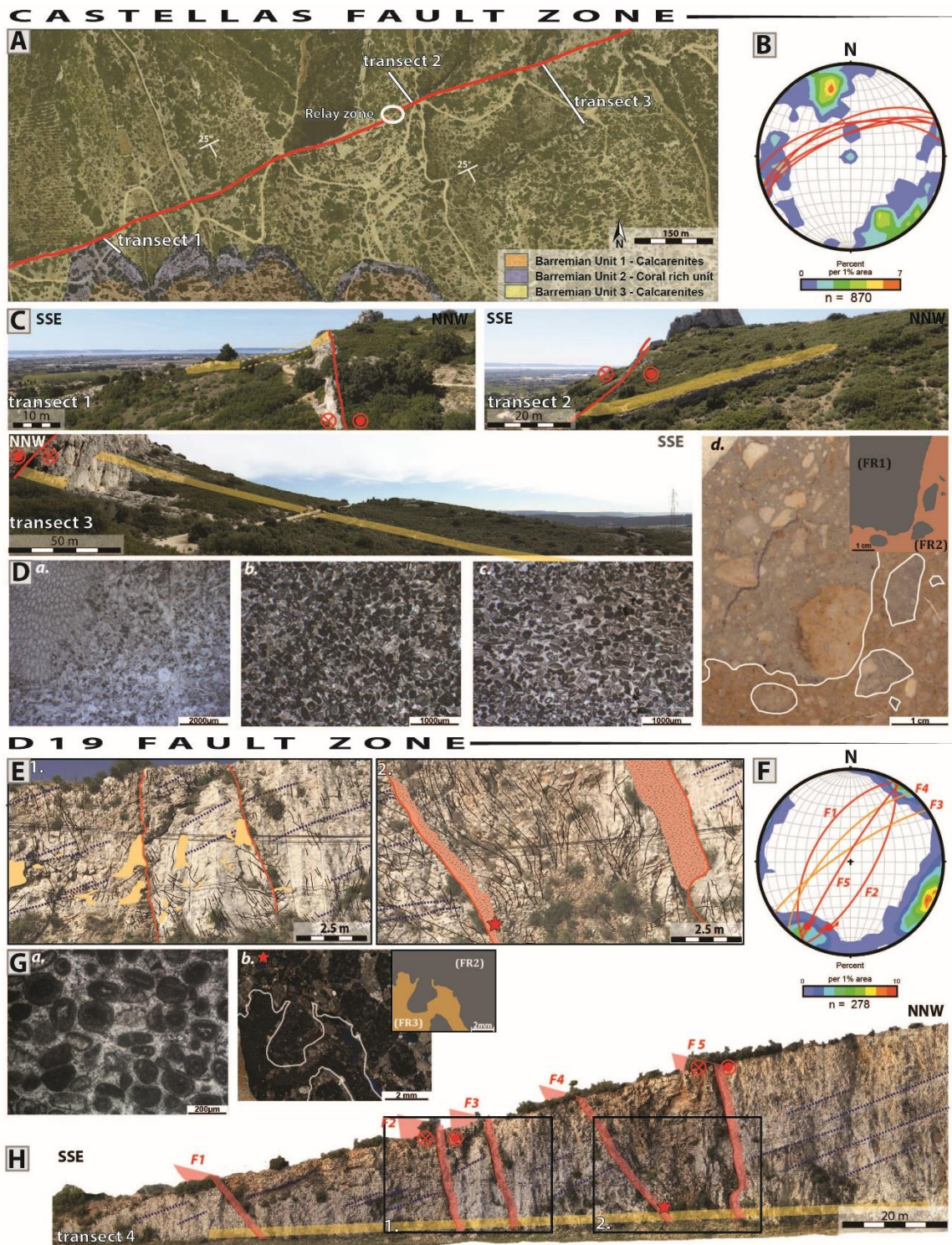


Figure 2 : A: Castellás fault map on aerial photo with localization of the studied transects and the relay zone; B: stereographic projections of poles to fractures (density contoured) and faults (red points) (Allmendinger et al., 2013; Cardozo and Allmendinger, 2013); C: Photos of transects; D: Carbonate host-rock facies (a) transect 1 coral rich unit, (b) transect 2 calcarenites, (c) transect 3 calcarenites and (d) fault rocks 1 and 2; E: Pictures of D19 outcrop F: Stereographic projections of poles to fractures (density contoured), set one faults (orange) and set 2 faults (red) G: Host rock facies (a) and of fault rocks (b); H: D19 outcrop including the five faults F1 to F5.

Table 1: structural properties of the fault zones

Faut zones	Faille	Direction	Dip	Dip direction	pitch striation	Fault core thickness	Fault Rocks		
							FR1	FR2	FR3
Castellas	Castellas	060 - 070	40 to 80	N	14W -	0 to 4m	sparsely present	majoritarily present	/
D19	F1	030	56	W		20	/	<10 cm	/
	F2	029	70	E	28 S	10 to 15	/	?	non constant thickness
	F3	056	80	N		0 to 15	/	?	?
	F4	042	70	W		20	/	in the clasts of FR3	non constant thickness
	F5	032	85	N	20 SW	50 to100	/	/	non constant thickness

3. DATA BASE

We performed 4 transects (T1 to T4) across the Castellas Fault and the D19 Fault (Fig. 2). Transect T1 is located along the coral rich unit 2. This bed is essentially composed of pelloidal grains and bioclasts (corals, bivalves and stromatoporidae; Fig. 2D a). Transects T2 and T3 are in unit 3, made of fine calcarenites with pelloidal grains and a rich fauna (foraminifera, bivalves, ostracods and echinoderm; Fig. 2Db, c). Transect 4 was conducted along the D19 outcrop (Fig. 3), which exposes Barremian outer platform bioclastic calcarenite with current ripples. The grains are mainly peloids with minor amount of bioclasts (solidary corals, bryozoan, bivalves and some rare miliolids; Fig. 2G, a).

The different tectonic events impacted the fault zone and fault core structure. Both faults have different fault cores (Table 1) made of 3 fault rock types in Castellas (Matonti et al., 2012) and D19 fault zones (see Aubert et al. 2019a). Fault rock 1 (FR1) results from the normal activation of the Castellas fault during Durancian uplift. It is a cohesive breccia composed of sub-rounded to rounded clasts from the nearby damage zone and <30% of grey matrix (Fig. 2Dd). Fault rock 2 (FR2), is linked to the sinistral reactivation of the Castellas fault and the onset of D19 fault zone during the Pyrenean shortening. FR2 present 2 morphologies depending on the fault zones. Within Castellas fault, FR2 is an un-cohesive breccia with an orange/oxidized matrix with angular to sub-rounded clasts from the damage zone and from FR1 (Fig. 2Dd). In the D19 fault zone, FR2 is a cohesive breccia with rounded clasts of the damage zone and a white cemented matrix (Fig. 2Gb). Fault rock 3 (FR3) is formed by the reactivation of D19 fault zone. It is composed of angular to sub-angular clast from FR2 and from the nearby damage zone in an orange/oxidized matrix (<20%) (Fig. 2Gb).

4. METHODS

The data set comprises 122 samples, 62 from Castellas and 60 from D19 outcrops, collected along the 4 transects. Porosity values were measured on 92 dry plugs with a Micromeritics AccuPyc 1330 helium pycnometer. Microfacies and petrography were determined on 92 thin sections. Impregnation with a blue-epoxy resin allowed us to decipher the different pore types. Thin sections were coloured with Alizarin red S and potassium ferricyanide to distinguish carbonate minerals (calcite and dolomite). The thin sections were analyzed using cathodoluminescence to discriminate the different calcite cements. The paragenetic sequence was defined based on superposition and overlap principles observed on thin sections using a Technosyn Cold Cathode Luminescence Model 8200 Mk II coupled to an Olympus_BH2 microscope and to a Zeiss_MR C5. Micrite micro-fabric and major element composition of 2

samples from the fault zone, 2 from the host rock and 1 from the D19 karst infilling were measured using PHILIPS XL30 ESEM with a current set at 20kV on fresh sample surface and on thin sections. To determine stable carbon and oxygen isotopes ($\delta^{13}\text{C}$ and $\delta^{18}\text{O}$), 204 microsamples (<5 mg) were drilled, 194 of them were micro-drilled from polished thin sections with an 80 μm diameter micro-sampler (Merkantec Micromill) at the VU University (Amsterdam, The Netherlands). We micro-sampled bulk rocks (57), sparitic cements (101), fault rocks (9) and micrite (27). Carbon and oxygen values were acquired with Thermo Finnigan Delta + mass spectrometer equipped with a GASBENCH preparation device at VU University Amsterdam. The internationally used standard IAEA-603, with official values of +2.46‰ for $\delta^{13}\text{C}$ and -2.37‰ for $\delta^{18}\text{O}$, is measured as a control standard. The SD of the measurements is respectively < 0.1‰ and < 0.2 ‰ for $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$. Ten whole rock samples were analysed using a Gasbench II connected to a Thermo Fisher Delta V Plus mass spectrometer at the FAU University (Erlangen, Germany). Measurements were calibrated by assigning $\delta^{13}\text{C}$ values of +1.95‰ to NBS19 and -47.3‰ to IAEA-CO9 and $\delta^{18}\text{O}$ values of -2.20‰ to NBS19. All values are reported in per mil relative to V-PDB.

5. RESULTS

1. MICROPOROSITY AND POROSITY

Porosity measurements performed on the 92 samples show that in average, porosity strongly decreases towards the fault core (Fig. 3): from more than 10% (mean: 15%, SD: 2.68 for

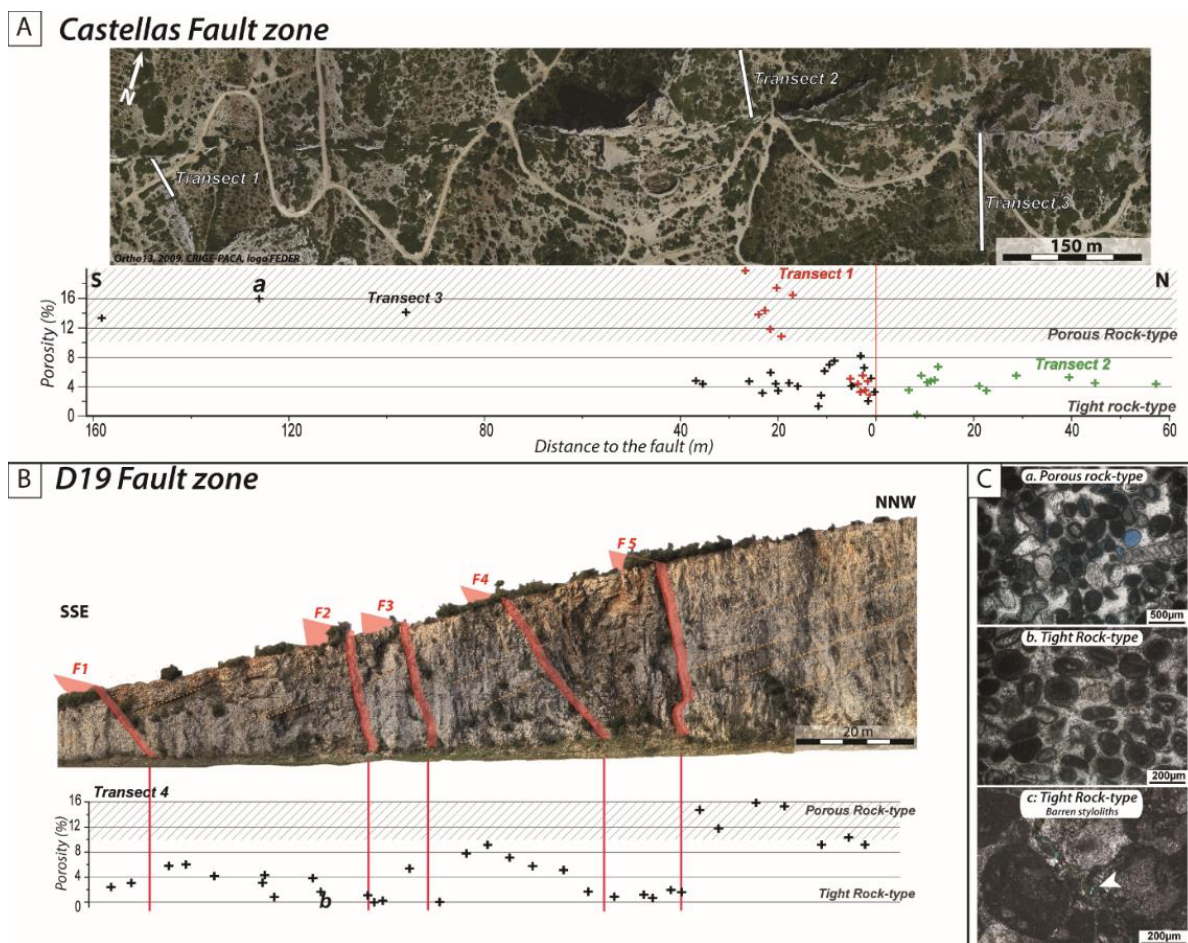


Figure 3: A: Castellas fault zone aerial view (Ortho13, 2009, CRIGE-PACA, logo FEDER) & porosity values measured along transect 1 (Red Cross), transect 2 (green cross) and transect 3 (black cross); B: porosity values measured along D19 fault zone; C: Pore types in the host rock (a) and in the fault zones (b&c).

Castellas and mean 12.3%, SD: 2.52 for D19) to less than 5% in fault zones (mean: 4.8%, SD: 2.07 for Castellas and mean: 3.16%, SD: 2.35 for D19).

Some variations occur as follows:

- North of the Castellas fault, along the 60m-long transect T2 the porosity is constantly low < 7% (mean of 4.4%, SD:1.53 ; Fig. 3A),
- South of the Castellas fault, the reduced porosity zone is >40m in transect 3 and 30m in transect 1 (Fig. 3A). In a 10m-thick zone from the fault plane, porosity reduction occurs with lower values in T1 (average 4.9%) than in T3 (average 5.6%).
- In the D19 fault zone, the lowest porosity values are in narrow zones around the faults (less than 2m) and in the lens between F4 and F5. Though, this porosity decrease is not homogeneous in fault zone and high values are found north of F1 and F3 (Fig. 3B).

From thin sections impregnated with blue-epoxy resin, a porous rock-type with $\phi > 10\%$ mainly in micritized grains as microporosity and moldic porosity (Fig. 3C a), and a tight rock-type with $\phi < 5\%$ where the porosity is mostly linked to barren styloliths (Fig. 3C b, c) are distinguished.

2. DIAGENETIC PHASES

a. Micrite micro-fabric

Micritized bioclasts, ooids and peloids were observed after SEM analysed of 2 fault zones samples and 2 host rock samples. Two micro-fabrics of micrite occur with specific crystal shape, sorting and contacts according to *Fournier et al.* (2011). Within both fault zones, the micrite is tight, with compact subhedral mosaic crystals (MF1; Fig. 4A, 4B). In the host rock, the micrite is loosely packed, and partially coalescent with puntic rarely serrate, subhedral to euhedral crystals (MF3; Fig. 4C, D, E). MF1 correlates with low porosity values < 5% , while MF3 with higher porosity > 10%.

b. Diagenetic cements

Eight cement stages were identified (Fig. 5). The red stain links to Alizarin red S coloration shows that all visible cements made up of calcite, which exhibits variable characteristics (morphology, luminescence, size and location).

The first 2 cement phases occur in both fault zones. The first cement (C0) is non-luminescent isopachous calcite of constant thickness ($\approx 10\mu\text{m}$) around grains (Fig. 5A). The second cement (C1) is divided in 2 sub-phases: a non-luminescent calcite, C1a, with a dog tooth morphology in intergranular spaces, and a bright luminescence calcite, C1b, covering C1a with a maximum thickness of $\approx 100\mu\text{m}$ (Fig. 5). C1b also fills micro-porosity in micritised grains (Fig. 5B). C1b values strongly increase in Castellas fault zone.

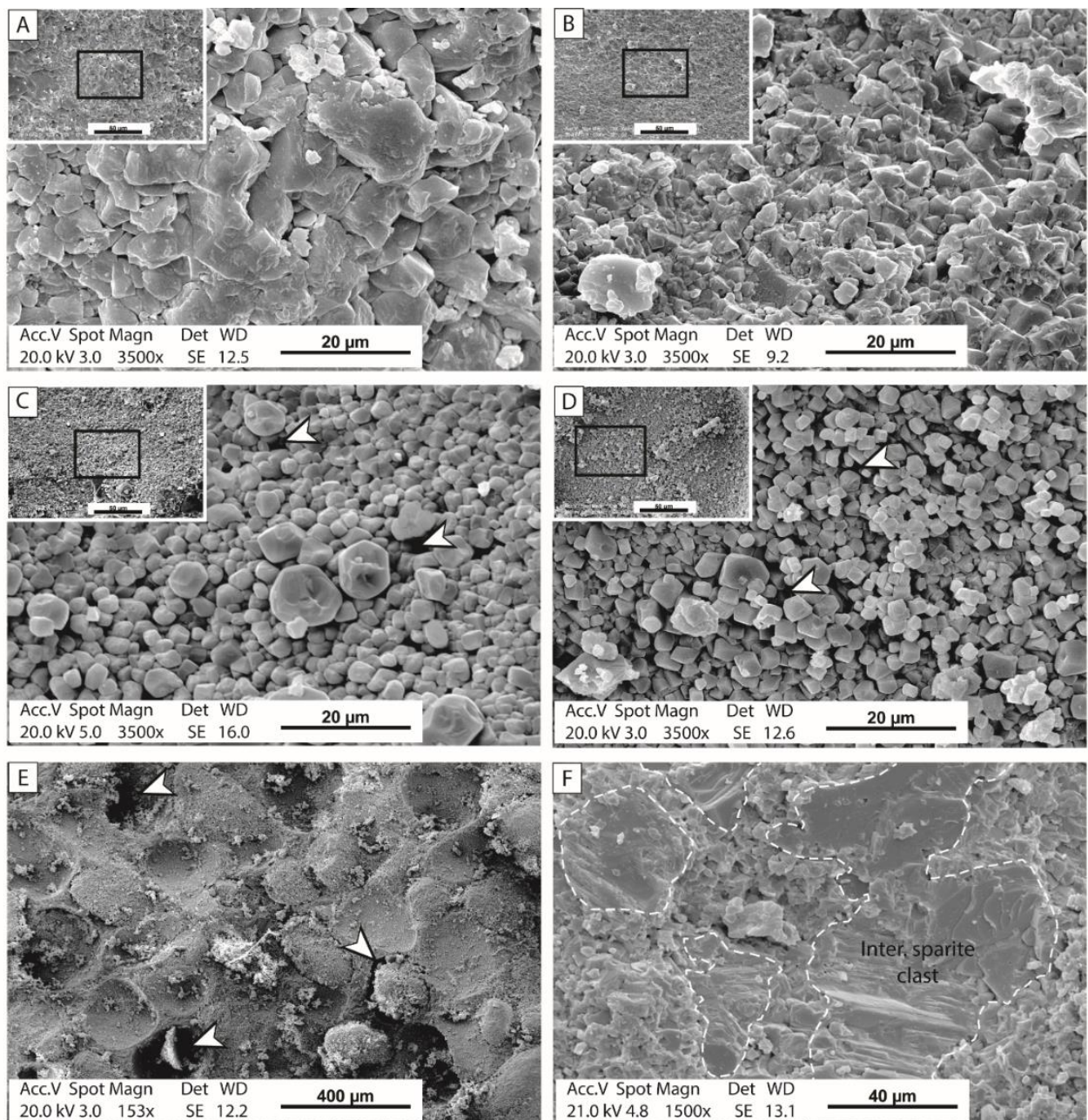


Figure 4 : MEB pictures of micrite micro-fabric and microporosity (white arrow); A. MF1micrite micro-fabric in Castellás fault zone (2.5m to fault plane); B. MF1micrite micro-fabric within D19 fault zones (2m away from F5 fault plane); C C: MF3 micrite micro-fabric within Castellás host rock (188m away from the fault plane); D: MF3 micrite micro-fabric within D19 host rock (95m away from F5 fault plane); E: D19 host rock moldic porosity; F. Karst infilling.

Five cements or replacive phases occur largely in the Castellás sector and rarely in the D19 outcrop:

- C2 is a sparitic cement with dull orange luminescence only found in fault core veins (Fig. 5B). SEM measurements show the Si and Al elements in the C2 veins. Most of Si crystals are an automorphic.
- C3 is a blocky calcite with non to red dull luminescence in veins, moldic and intergranular pores (Fig. 5B, C, D). This cement also occurs in few veins of D19 sectors but is not restricted to the fault zone.

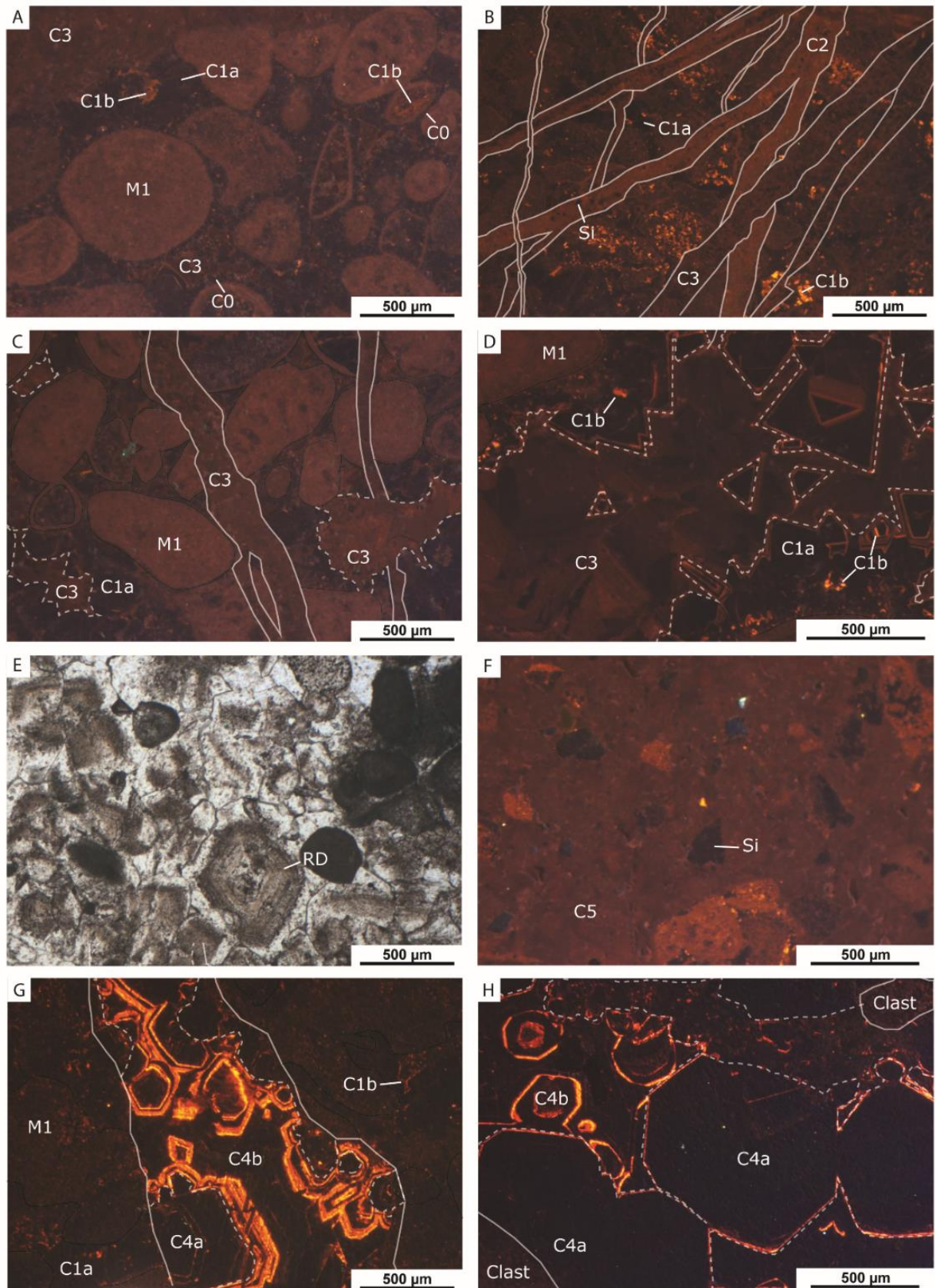


Figure 5 : Thin-sections under cathodoluminescence; A: Calcarenite in transect 3 with micritized grain (M1), and intergranular space cemented with C1 a&b and C3; B: C2 (with Si) and C3 veins affecting Castellás FR1 clast with micritized grains cemented by C1b; C: C3 vein cement and intergranular space in Castellás fault zone; D: C1 (a & b) and C3 cementing moldic porosity of transect 3 calcarenite; E: FR1 matrix with phantom of cloudy appearance replacive dolomite; F: FR1 matrix de-dolomitized by C5 containing quartz grains; G: C4 (a & b) cementing vein of D19 fault zone; H: matrix of D19 FR2 cemented by C4 (a&b).

- Phantoms of planar-e (euhedral) dolomite crystals (Sibley and Gregg, 1987) with a maximum size of 500µm affect the matrix of FR1 (Fig. 5E). They are vestiges of a dolomitization phase. They have a cloudy appearance caused by solid micritic inclusion in the crystal and can be considered as replacive dolomite (RD; Machel, 2004). Within the FR1 matrix, an important concentration of angular grains of quartz with a maximum size of 300µm is noticed (Fig. 5F).
- A blocky calcite C4 (referred to as S2 in Aubert et al. (2019a)) is mainly present in veins of the D19 outcrop, in matrix of FRA, and intergranular and moldic pores (Fig. 5G, 5H). This cement shows zonation of non-luminescent and bright luminescent bands and can be divided in 2 sparitic sub-phases: C4a which is non-luminescent with some highly luminescent band and C4b which is bright luminescent with some non-luminescent bands. C4a occurs in lesser proportion in some veins along transect T2 and T3 of the Castellás fault.
- A sparitic cement C5, with a red dull luminescence replaces the RD phase (Fig. 5F).

c. Additional diagenetic features

In addition to cementation phases, other diagenetic elements affected both fault zones. Karst infilling occurs in the F2 fault zone of the D19 outcrop. It is composed of well-sorted grains deposited in laminated layers. This formation presents a stack of micrite-rich layers and grain-rich layers. In which grains are intergranular sparitic clasts, remaining from blocky calcite of dissolved grainstones and oxydes. The laminated layers are affected by veins and stylolites; some of these are deformed due to the clasts fall on sediments. Micritic layers has been observed under SEM, the micrite appeared tight with compact subhedral mosaic crystals (Fig. 4F). We observed oxide filling mainly in the Castellás area in dissolution voids affecting C1a, C1b and C3 cementation phases and in D19 in karstic fill. The proportion of oxides increase close to stylolites.

3. CARBON AND OXYGEN ISOTOPES

Isotope measurements were realized on samples collected along transects of the fault zones. A hundred and eighty-nine measurements of C and O isotopes were performed on 16 samples and 32 thin sections (Fig. 6A, table 2).

Sampling was done in bulk rock (66), sparitic cement (101; veins, intergranular spaces and fault rock cements) and in fault rocks (10) in order to determine their isotopic signature. Isotopic values range from -10.40‰ to -3.65‰ for $\delta^{18}\text{O}$ and from -7.20‰ to +1.42‰ for $\delta^{13}\text{C}$ (Fig. 6A, 6B, table 2). The bulk rock values range from -9.18‰ to -4.34‰ for $\delta^{18}\text{O}$ and from -4.80‰ to +1.19‰ for $\delta^{13}\text{C}$ (Fig. 6A, table 2). These values are split in 2 sets. Set one includes transect 1 & 3 of the Castellás Fault. Bulk values range from -6.07‰ to -4.34‰ for $\delta^{18}\text{O}$ and from -1.41‰ to +1.19‰ for $\delta^{13}\text{C}$. Set 2 includes transect 2 (Castellás) and transect 4 (D19). Bulk values range from -9.18‰ to -5.20‰ for $\delta^{18}\text{O}$ and from -4.80‰ to -0.60‰ for $\delta^{13}\text{C}$ (Fig. 6B, table 2). In the transect 3, the isotopic values only slightly vary along transect, ranging from -6.13‰ to -4.50‰ for $\delta^{18}\text{O}$ and from -1.41‰ to +0.47‰ for $\delta^{13}\text{C}$ (Fig. 6C, table 2). Contrarily, values vary more along the D19 transect. They range from -9.18‰ to -5.20‰ for $\delta^{18}\text{O}$ and from -4.80‰ to -0.60‰ for $\delta^{13}\text{C}$ (Fig. 6C, table 2). Indeed, the $\delta^{13}\text{C}$ values obviously decrease in the fault vicinity, especially south of F2.

Isotopic values of cements filling veins, intergranular spaces, karst fills, and fault rock are divided into 5 groups (Fig. 6A, table 2):

- the group of values from C1 fluctuates from -6.76‰ to -4.45‰ for $\delta^{18}\text{O}$ and from -1.28 to +1.08‰ for $\delta^{13}\text{C}$;
- the group of values from C3 ranges from -10.40‰ to -6.73‰ for $\delta^{18}\text{O}$ and from -2.09 to +1.22‰ for $\delta^{13}\text{C}$;
- the group of values of C4 in FR1 and FR2 matrix and in karst fill ranges from -9.18‰ to -4.60‰ for $\delta^{18}\text{O}$ and from -5.10‰ to -0.74‰ for $\delta^{13}\text{C}$ with a positive covariance between $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$. FR 2 matrix values (from -6.55 to -7.06‰ for $\delta^{18}\text{O}$ and from -1.10 to -2.24‰ for $\delta^{13}\text{C}$) present slightly less negative values than karst fill with mean values of -7.83‰ and -2.53‰ respectively for $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$. (Fig. 6A). In the Castellás fault, 4 isotopic values from 2 veins are high with means of -6.25 and -4.2‰ for $\delta^{18}\text{O}$ - 0.64 and -0.09‰ for $\delta^{13}\text{C}$ having similar positive covariance than the other C4 values.
- the group of values from C5, sampled in FR1 matrix with a mean of -7.49‰ for $\delta^{18}\text{O}$ and -4.01‰ for $\delta^{13}\text{C}$ (Fig. 6A).
- the group of values from FR3 matrix with a mean of -5.98‰ for $\delta^{18}\text{O}$ and -6.83‰ for $\delta^{13}\text{C}$ (Fig. 6A)

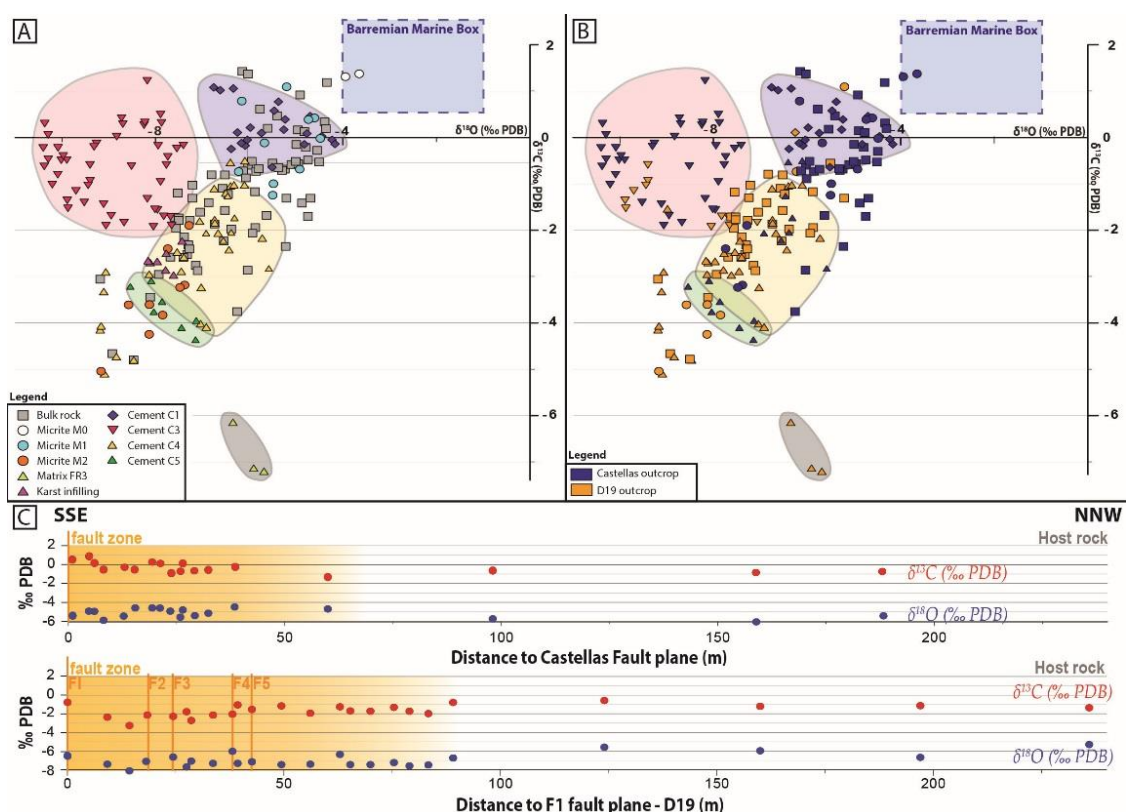


Figure 6 : Isotopic values of $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ measured on bulk rock, cement phases, and micrite. Range values of “Urgonian marine box” from Moss & Tucker (1995) and Godet et al. (2006); A: set of values sorted by the nature of diagenetic phases and B: values sorted by the fault zone; C: lateral evolution of $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ isotopic values in Castellás (top) and in D19 (bottom) fault zones.

270 **Table 2:** Carbon and oxygen isotope values of bulk carbonates for Castellás fault zone and D19
271 fault zones. B: bulk measurements; M: micrite values; C1, C3, C4, C5: isotopic values of
272 cement C1, C3, C4 and C5; FR: fault rock isotopic values.

Transect	Sample	$\delta^{13}\text{C}$ (‰ vs VPDB)	$\delta^{18}\text{O}$ (‰ vs VPDB)	Class	Distance to F. (m)
Castellás (T 1)	201	1,19	-4,34	B	1,3
Castellás (T 1)	201	1,02	-6,62	C1	1,3
Castellás (T 1)	201	1,31	-3,94	M	1,3
Castellás (T 1)	201	1,37	-3,65	M	1,3
Castellás (T 1)	213	-0,68	-5,24	B	22,7
Castellás (T 1)	213	-0,58	-5,10	B	22,7
Castellás (T 1)	213	-0,18	-6,09	C1	22,7
Castellás (T 1)	213	0,03	-4,45	C1	22,7
Castellás (T 1)	213	0,09	-4,77	C1	22,7
Castellás (T 1)	213	-2,09	-6,92	C4	22,7
Castellás (T 1)	213	-0,68	-4,92	M	22,7
Castellás (T 2)	c3b17	-0,52	-5,95	B	4,6
Castellás (T 2)	c3b17	-2,07	-6,38	C4	4,6
Castellás (T 2)	c3b7	-0,64	-5,51	B	9,3
Castellás (T 2)	c3b26	-3,76	-6,26	B	22,6
Castellás (T 2)	c3b26	-2,85	-5,58	C4	22,6
Castellás (T 2)	c3b26	-1,31	-4,69	B	57,3
Castellás (T 2)	c3b7	-1,76	-6,31	C1	57,3
Castellás (T 2)	c3b7	-1,28	-6,46	C1	57,3
Castellás (T 2)	c3b26	-2,35	-5,22	M	57,3
Castellás (T 2)	c3b26	-1,70	-4,75	M	57,3
Castellás (T 3)	327	-0,24	-7,55	C3	0,3
Castellás (T 3)	325	-1,90	-9,06	C3	0,3
Castellás (T 3)	325	-1,69	-8,95	C3	0,3
Castellás (T 3)	327	-3,11	-8,09	C4	0,3
Castellás (T 3)	327	0,47	-5,40	B	1,0
Castellás (T 3)	327	-0,18	-7,95	C3	1,0
Castellás (T 3)	327	-0,17	-7,41	C3	1,0
Castellás (T 3)	328	0,10	-5,74	C1	1,6
Castellás (T 3)	328	-1,32	-8,18	C3	1,6
Castellás (T 3)	328	-0,59	-7,77	C3	1,6
Castellás (T 3)	328	-0,42	-7,74	C3	1,6
Castellás (T 3)	328	-0,13	-9,26	C3	1,6
Castellás (T 3)	328	0,02	-8,83	C3	1,6
Castellás (T 3)	328	0,29	-8,70	C3	1,6
Castellás (T 3)	328	0,42	-8,73	C3	1,6
Castellás (T 3)	328	0,50	-7,89	C3	1,6
Castellás (T 3)	328	1,22	-8,18	C3	1,6
Castellás (T 3)	333	-1,84	-8,67	C3	1,6

Castellas (T 3)	333	-0,96	-7,89	C3	1,6
Castellas (T 3)	328	-0,14	-4,17	C4	1,6
Castellas (T 3)	328	-0,05	-4,23	C4	1,6
Castellas (T 3)	329	0,16	-4,95	B	2,4
Castellas (T 3)	333	-0,25	-6,38	C1	4,6
Castellas (T 3)	333	-0,12	-6,17	C1	4,6
Castellas (T 3)	333	-0,62	-8,52	C3	4,6
Castellas (T 3)	333	-0,12	-5,67	M	4,6
Castellas (T 3)	333	-0,02	-4,48	M	4,6
Castellas (T 3)	333	0,42	-4,60	M	4,6
Castellas (T 3)	337	0,19	-5,59	B	9,5
Castellas (T 3)	302	-0,53	-4,50	B	11,8
Castellas (T 3)	302	-0,49	-4,74	B	11,8
Castellas (T 3)	302	-0,62	-10,38	C3	11,8
Castellas (T 3)	302	-0,49	-10,02	C3	11,8
Castellas (T 3)	305	0,33	-4,38	B	16,0
Castellas (T 3)	306	0,21	-4,35	B	17,8
Castellas (T 3)	307	-0,01	-4,46	B	18,2
Castellas (T 3)	308	-0,57	-4,95	B	20,0
Castellas (T 3)	308	-1,44	-9,11	C3	20,0
Castellas (T 3)	308	-0,23	-10,40	C3	20,0
Castellas (T 3)	308	-0,22	-10,08	C3	20,0
Castellas (T 3)	309	-1,41	-4,87	B	20,5
Castellas (T 3)	309	-0,52	-5,01	B	20,5
Castellas (T 3)	309	-0,15	-4,82	C1	20,5
Castellas (T 3)	309	-1,56	-7,96	C3	20,5
Castellas (T 3)	309	-1,55	-8,01	C3	20,5
Castellas (T 3)	312	0,12	-4,81	B	23,2
Castellas (T 3)	314	-0,71	-5,30	B	25,9
Castellas (T 3)	314	-0,80	-10,09	C3	25,9
Castellas (T 3)	314	-0,49	-9,90	C3	25,9
Castellas (T 3)	314	-0,47	-10,29	C3	25,9
Castellas (T 3)	314	-0,40	-9,97	C3	25,9
Castellas (T 3)	314	0,06	-10,30	C3	25,9
Castellas (T 3)	316	-1,24	-5,50	B	29,2
Castellas (T 3)	316	-1,00	-5,48	B	29,2
Castellas (T 3)	316	-0,22	-4,79	B	29,2
Castellas (T 3)	316	-1,02	-10,21	C3	29,2
Castellas (T 3)	316	-0,18	-9,31	C3	29,2
Castellas (T 3)	316	0,30	-10,37	C3	29,2
Castellas (T 3)	318	-0,28	-4,53	B	35,4
Castellas (T 3)	320	-0,68	-5,79	B	96,1
Castellas (T 3)	322	-0,88	-6,07	B	158,0
Castellas (T 3)	323	-0,65	-5,37	B	188,0
Castellas (ZF1)	Z1,1	0,17	-5,26	C1	0,0
Castellas (ZF1)	Z1,1	0,39	-5,23	C1	0,0

Castellas (ZF1)	Z1,1	0,46	-4,70	C1	0,0
Castellas (ZF1)	Z1,2	0,21	-5,98	C1	0,0
Castellas (ZF1)	Z1,1	-0,55	-6,40	C4	0,0
Castellas (ZF1)	Z1,1	-0,52	-6,10	C4	0,0
Castellas (ZF1)	Z1,2	-4,12	-7,45	C5	0,0
Castellas (ZF1)	Z1,2	-0,15	-4,99	FR	0,0
Castellas (ZF1)	Z1,2	0,39	-4,73	M	0,0
Castellas (ZF1)	Z1,2	0,61	-5,77	M	0,0
Castellas (ZF1)	Z1,1	0,78	-6,16	M	0,0
Castellas (ZF2)	Z2,2	0,77	-5,38	C1	0,0
Castellas (ZF2)	Z2,7	-1,40	-9,52	C3	0,0
Castellas (ZF2)	Z2,7	-4,38	-7,15	C5	0,0
Castellas (ZF2)	Z2,7	-3,97	-7,13	C5	0,0
Castellas (ZF2)	Z2,7	-3,78	-8,04	C5	0,0
Castellas (ZF2)	Z2,7	-3,56	-7,86	C5	0,0
Castellas (ZF2)	Z2,7	-3,24	-7,48	C5	0,0
Castellas (ZF2)	Z2,7	-3,23	-8,54	C5	0,0
Castellas (ZF2)	Z2,2	0,58	-5,47	FR	0,0
Castellas (ZF2)	Z2,2	0,92	-4,91	FR	0,0
Castellas (ZF2)	Z2,7	-1,68	-5,63	FR	0,0
Castellas (ZF2)	Z2,7	-2,24	-6,55	FR	0,0
Castellas (ZF2)	Z2,7	-3,18	-7,38	M	0,0
Castellas (ZF2)	Z2,7	-2,86	-6,03	FR	1,0
Castellas (ZF5)	Z5,4	0,27	-8,25	C3	0,0
Castellas (ZF5)	Z5,4	0,31	-7,87	C3	0,0
Castellas (ZF5)	Z5,4	0,32	-8,23	C3	0,0
Castellas (ZF5)	Z5,4	1,06	-6,34	C1	0,4
Castellas (ZF5)	Z5,4	1,08	-6,76	C1	0,4
Castellas (ZF5)	Z5,4	1,05	-7,13	FR	0,4
Castellas (ZF5)	Z5,4	1,37	-6,03	FR	0,4
Castellas (ZF5)	Z5,4	1,42	-6,15	FR	0,4
Transect	Sample	$\delta^{13}\text{C}$ (‰ vs VPDB)	$\delta^{18}\text{O}$ (‰ vs VPDB)	Class	Distance to F1 (m)
D19	3B	-0,81	-6,52	B	0,0
D19	3B	-1,20	-6,50	C1	0,0
D19	3B	-1,02	-6,33	C1	0,0
D19	3B	0,11	-6,25	C1	0,0
D19	3B	-0,74	-6,23	M	0,0
D19	9	-2,32	-7,30	B	9,2
D19	13a	-3,44	-8,11	B	14,3
D19	13a	-2,96	-7,93	B	14,3
D19	13C	-2,97	-7,62	M	14,3
D19	13C	-2,86	-7,79	M	14,3
D19	13C	-2,70	-8,12	M	14,3
D19	13C	-2,67	-7,96	M	14,3

D19	13C	-2,66	-8,16	M	14,3
D19	13C	-2,50	-7,77	M	14,3
D19	13C	-1,54	-8,98	M	14,3
D19	17	-2,58	-7,68	B	18,7
D19	14A	-1,97	-6,38	B	18,7
D19	14A	-1,87	-6,74	B	18,7
D19	15B	-2,23	-7,43	B	18,7
D19	17	-1,05	-6,40	C1	18,7
D19	14A	-1,77	-6,74	C1	18,7
D19	14A	-2,42	-6,43	C4	18,7
D19	14A	-2,06	-6,67	C4	18,7
D19	21	-2,23	-6,54	B	24,4
D19	RSG	-1,90	-7,66	B	28,4
D19	RSG	-1,70	-7,83	B	28,4
D19	RSD	-2,87	-7,10	B	29,5
D19	RSD	-2,76	-7,14	B	29,5
D19	RSD	-0,93	-9,40	C3	29,5
D19	RSF1	-2,40	-7,28	B	34,7
D19	RSF2	-2,14	-7,39	B	34,7
D19	RSF2	-1,78	-7,27	B	34,7
D19	RSF1	-1,03	-9,44	C3	34,7
D19	RSF2	-1,93	-8,05	C3	34,7
D19	RSF2	-0,59	-9,40	C3	34,7
D19	RSF2	-2,95	-8,14	C4	34,7
D19	RSE 1	-2,53	-7,33	B	35,0
D19	RSE 2	-2,59	-7,41	B	35,0
D19	RSE 1	-1,71	-7,68	C3	35,0
D19	RSE 2	-1,84	-6,73	C3	35,0
D19	57	-2,07	-5,93	B	38,1
D19	57	-1,94	-5,87	B	38,1
D19	57	-1,83	-7,06	C3	38,1
D19	57	-1,10	-6,75	C3	38,1
D19	57	-4,02	-7,04	C4	38,1
D19	57	-2,17	-5,72	C4	38,1
D19	57	-1,58	-6,52	FR	38,1
D19	57	-7,20	-5,68	M	38,1
D19	57	-7,13	-5,90	M	38,1
D19	28b	-1,03	-7,21	B	39,3
D19	28b	-1,03	-6,10	C3	39,3
D19	28b	-4,09	-6,92	C4	39,3
D19	28b	-2,58	-7,40	C4	39,3
D19	28b	-2,47	-7,54	C4	39,3
D19	30a	-1,61	-7,04	B	42,6
D19	30a	-1,41	-6,87	B	42,6
D19	30a	-3,23	-7,03	C4	42,6
D19	30a	-2,89	-7,45	C4	42,6

D19	24a	-1,21	-7,52	B	51,1
D19	27b	-1,92	-7,48	B	57,9
D19	31	-1,24	-6,44	B	65,0
D19	32	-1,75	-7,50	B	67,4
D19	34	-1,79	-7,49	B	72,2
D19	36	-1,32	-7,21	B	77,8
D19	38	-1,73	-7,59	B	81,5
D19	62	-1,96	-7,56	B	86,0
D19	42	-0,81	-6,80	B	91,9
D19	63	-0,55	-5,50	B	124,0
D19	64	-1,17	-5,88	B	160,0
D19	65	-1,10	-6,57	B	197,0
D19	66	-1,31	-5,21	B	236,0
D19	60a	-3,06	-9,18	B	255,2
D19	60B	-4,80	-8,47	B	255,2
D19	60B	-4,66	-8,92	B	255,2
D19	61	-1,53	-9,87	C3	255,2
D19	61	-1,36	-9,89	C3	255,2
D19	60a	-1,15	-9,70	C3	255,2
D19	60a	-3,32	-9,11	C4	255,2
D19	60B	-5,10	-9,09	C4	255,2
D19	60B	-4,73	-8,84	C4	255,2
D19	60B	-4,15	-9,18	C4	255,2
D19	60B	-4,07	-9,16	C4	255,2
D19	60B	-2,90	-9,06	C4	255,2
D19	60a	-3,83	-7,85	M	255,2
D19	60B	-5,04	-9,17	M	255,2
D19	60B	-4,25	-8,14	M	255,2
D19	60B	-3,61	-8,58	M	255,2
D19	60B	-3,61	-8,13	M	255,2

6. DISCUSSION

1. DIAGENETIC EVOLUTION OF THE FAULT ZONES

The chronological relations between cements can be established thanks to cross-cutting relation and inclusion principles. Indeed, the veins filled with cement C2 cross-cut cements C1a and C1b (Fig. 5B). Thus, C2 cementation postponed C1 cementation. The C3 veins cross-cut the C2 veins, but are included within FR1 clasts (Fig. B). Hence, C3 cement is ante-FR1 development but post-C2 cementation. The fault rock 1 (FR1) is related to the first normal fault activity, consequently, C1, C2 and C3 cementation phases occurred prior to the proper fault plane and fault core formation and, are related to the fault nucleation. Replacive dolomite is within FR1 matrix (Fig. 3E), therefore, it develop after FR1 formation. Finally, the cement C4 can be noticed within FR2 matrix indicating that C4 cementation event postponed FR2 formation. The fault rock 2 (FR2) developed during the faults strike-slip reactivation. The combined superposition, overlap, cross-cutting principles and isotopic signature of cements brought out the chronology between phases, and revealed the paragenetic sequence (Fig. 7).

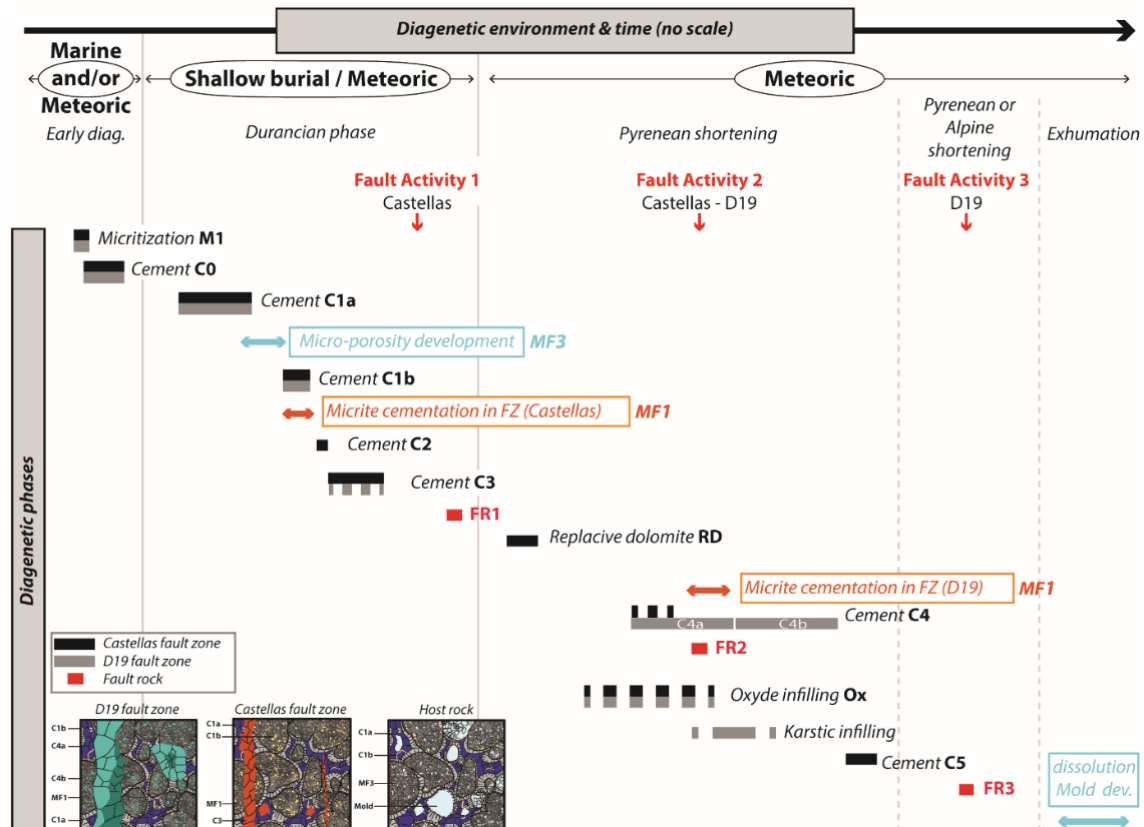


Figure 7: Paragenetic sequence of the both fault zones (black: Castellas, grey: D19) with micro-porosity development (blue) and cementation (orange) and fault zone activation (red).

The Urgonian carbonates in La Fare anticlinal underwent 3 important diagenetic events, which impacted the host rock and/or the fault zones. We discriminate among diagenetic events that occurred before and during faulting.

a. Pre fault diagenesis – microporosity development

During Upper Barremian, just after deposition, micro-bores organisms at the sediment-water interface enhanced the formation of micritic calcitic envelopes on bioclasts, ooids and peloids (Purser, 1980; Reid and Macintyre, 2000; Samankassou et al., 2005; Vincent et al., 2007). This micritisation in marine conditions is typical for Urgonian low energy inner platform (Fournier et al., 2011; Masse, 1976). Subsequently, cement C0 formed around grains and formed a solid shelf inducing the conservation of the clast shape during the later burial compaction (Step 0 on Fig. 8). However, the majority of isotopic values do not fit in the Barremian sea water calcite box which ranges from -1.00‰ to -4.00‰ for $\delta^{18}\text{O}$ and from +1.00‰ to +3.00‰ for $\delta^{13}\text{C}$ (Fouke et al., 1996; Godet et al., 2006). Only 2 data points sampled of micritised grains show isotopic values close the Barremian sea water calcite. The depletion of other data indicates the slight impact of C0 cementation on isotopic values.

The next sub-phase of cementation C1a partly fills intergranular porosity. This non luminescent cement with isotopic values ranging from -6.8‰ to -3.9‰ for $\delta^{18}\text{O}$ and from -1.0‰ to +1.3‰ for $\delta^{13}\text{C}$ is characteristic for mixed fluids. Léonide et al. (2014) measured a calcite cement S1, near La Fare anticline with similar luminescence and isotopic range values (mean: $\delta^{18}\text{O}$ = -5.49‰; $\delta^{13}\text{C}$ = +2.34‰). These authors linked this cementation phase to a shallow burial meteoric flow under equatorial climate during Durancian uplift. This diagenetic event led to micrite re-crystallization, and development of microporosity (MF3). Since La Fare carbonates

were exhumed at that time (Léonide et al., 2014) the meteoric fluids led to similar diagenetic modifications (Step 1 on Fig. 8):

- (i) Cementation of C1a, partly filling intergranular porosity (Fig. 9B1a)
- (ii) Micrite re-crystallization and microporosity MF3 setup by Ostwald ripening processes (Ostwald, 1886; Volery et al., 2010).

The micrite re-crystallization strongly increased rock porosity due to enhanced microporosity (Fig. 9B1b). Microporous limestones have a high matrix porosity but low to moderate matrix permeability (Deville de Periere et al., 2011; Jack and Sun, 2003). Indeed, in the case of Barremian limestones of La Fare anticline, porosity is >10% but located in the grains, what restricts possible flow pathways. Resulting from this event, Urgonian carbonates formed a type III reservoir *sensu* Nelson (2001).

b. Fault related diagenesis – alteration of reservoir properties

Normal faulting-related diagenesis

The Castellás fault first nucleated during Durancian uplift (Aubert et al., 2019b; Matonti et al., 2012) impacting the host Urgonian carbonates.

In porous granular media, fault nucleation mechanisms can lead to dilation processes (Fossen and Bale, 2007; Fossen and Rotevatn, 2016; Main et al., 2000; Wilkins et al., 2007; Zhu and Wong, 1997), and under low confining pressure (<100KPa; Alikarami & Torabi 2015). Because this process leads to dilatancy, it increases the rock permeability (Alikarami and Torabi, 2015; Bernard et al., 2002) in the first stage of deformation bands (Heiland et al., 2001; Lothe et al., 2002) what allows fluids to flow.

Castellás fault zone nucleated within a partially and dimly cemented host rock under low confining pressure, in an extensional stress pattern, at a depth <1km (Lamarche et al. 2012). Barremian host rock presented properties (porosity/stress pattern/confining pressure) close to the porous granular described above. Moreover, Micarelli et al. (2006) showed that, during early stages, fault zones in carbonates have a hydraulic behaviour comparable to deformation bands. Hence, in the Urgonian carbonates of La Fare sector, dilatant processes occurred as an incipient fault mechanism and enhanced fluid circulations along the deformation bands. These fluid flows led to the cementation of C1b (Step 2 on Fig. 8). However, dilation bands are unstable and grain collapse occurs swiftly after the beginning of the deformation due to an increase in the loading stresses (Lothe et al., 2002). This explains why C1b does not fill all intergranular porosity. Consequently, as all micritic grains in fault zone are cemented by C1b, the bulk isotopic measurements are strongly influenced by C1 cement isotopic values. This is the explanation why in transect 3 the bulk isotopic values 30m apart from the fault (means of -5.26‰ for $\delta^{18}\text{O}$ and -0.82‰ for $\delta^{13}\text{C}$) are close to bulk isotopic values far from the fault plane (188m; -5.37‰ for $\delta^{18}\text{O}$ and -0.65‰ for $\delta^{13}\text{C}$, Fig. III 6A). Others dilation bands has also been described by Kaminskaite et al. (2019) in the San Vito Lo Capo carbonates grainstones (Sicily, Italy). These dilation bands also led to cementation of the carbonate rock.

The C1a and C1b led to a local rock embrittlement and to a porosity decrease by cementation of the microporosity. During the first stages of fault evolution in low porosity limestones, intense fracturing of the fault zone predating fault core formation is known to increase the permeability (Micarelli et al., 2006). In the studied faults, the first brittle event allowed an Al-

rich fluid to flow with micro-metric quartz grains in the barren fractures, and C2 to cement (Step 3 on Fig. 8). The Urgonian facies of the studied area are composed of pure carbonates without siliciclastic input. Quartz grains and Aluminium could have been reworked from surrounding formations. The rocks underlying the studied exposed Urgonian carbonates are limestones and dolostones. Albian and Aptian rocks are marly and sandy limestones, respectively (Anglada et al., 1977). Hence, Aptian layers are very likely to be the source of quartz. The fluids must have carried small grains of quartz from the Aptian sandy limestones via the fracture network. The Al enrichment of C2 could result from the erosion of Albian and Aptian deposits during the Durancian uplift (Guendon and Parron, 1985; Triat, 1982).

As the fault zone grew, a new fracture set formed, leading to new phase of calcite cementation (C3) in veins and intergranular porosity (Step 4 on Fig. 8). The $\delta^{18}\text{O}$ isotopic values of C3 range from -10.40‰ to -6.73‰ with $\delta^{13}\text{C}$ values between -2.09‰ and +1.22‰. As C3 cementation occurred during the Durancian uplift and denudation, C3 most probably did not cement at high depth (depth of maximum 500m; Fig. 9C4). The negative $\delta^{13}\text{C}$ values tend corroborate that it would rather be a meteoric fluid than a marine fluid. Hence, C3 would correspond to a shallow burial/meteoric cementation phase. Resulting from this cementation, rocks in this zone tightened down to <5%. The porosity did not change since this event (Fig. 9 B5). This porosity reduction due to cementation has also been observed in other cases of brittle-dilatant faults (Agosta et al., 2007; Celico et al., 2006; Gaviglio et al., 2009; Mozley and Goodwin, 1995). Implicitly, the fault zone was a barrier to fluid flow, leading to a reservoir compartmentalization. The C3 fluid flow also occurred along fracture clusters of the D19 sector and led to vein formation.

In a later stage, the fault core formed and the fault plane *sensu-stricto* developed, leading to FR1 breccia with a permeable matrix with quartz grains >100µm in size (Step 5 on Fig. 8). These grains either came from silica from C2 in veins describe above or from Aptian overlying rocks. C2 silica crystals in veins are scarce and smaller than 10µm. Thus, quartz grains may rather come from Aptian rocks like the quartz found in C2 veins. The presence of Aptian quartz in the fault core proves that the Castellans fault affected Aptian rocks, which were later eroded during the Durancian uplift. Implicitly, the fault activity occurred before total erosion of Aptian rocks. Uncemented breccias within the fault core form good fluid pathways (Billi et al., 2008; Delle Piane et al., 2016). In the studied fault, formation of FR1 breccia allowed the fault core to act as a drain. However, the cemented surrounding host rocks constrained the drainage area of this high permeable conduit. Un-cemented breccias acting as good across- and along- fluid pathways were also described on Apennines carbonate formations within strike and normal faults fault core (Billi et al., 2003, 2008; Storti et al., 2003).

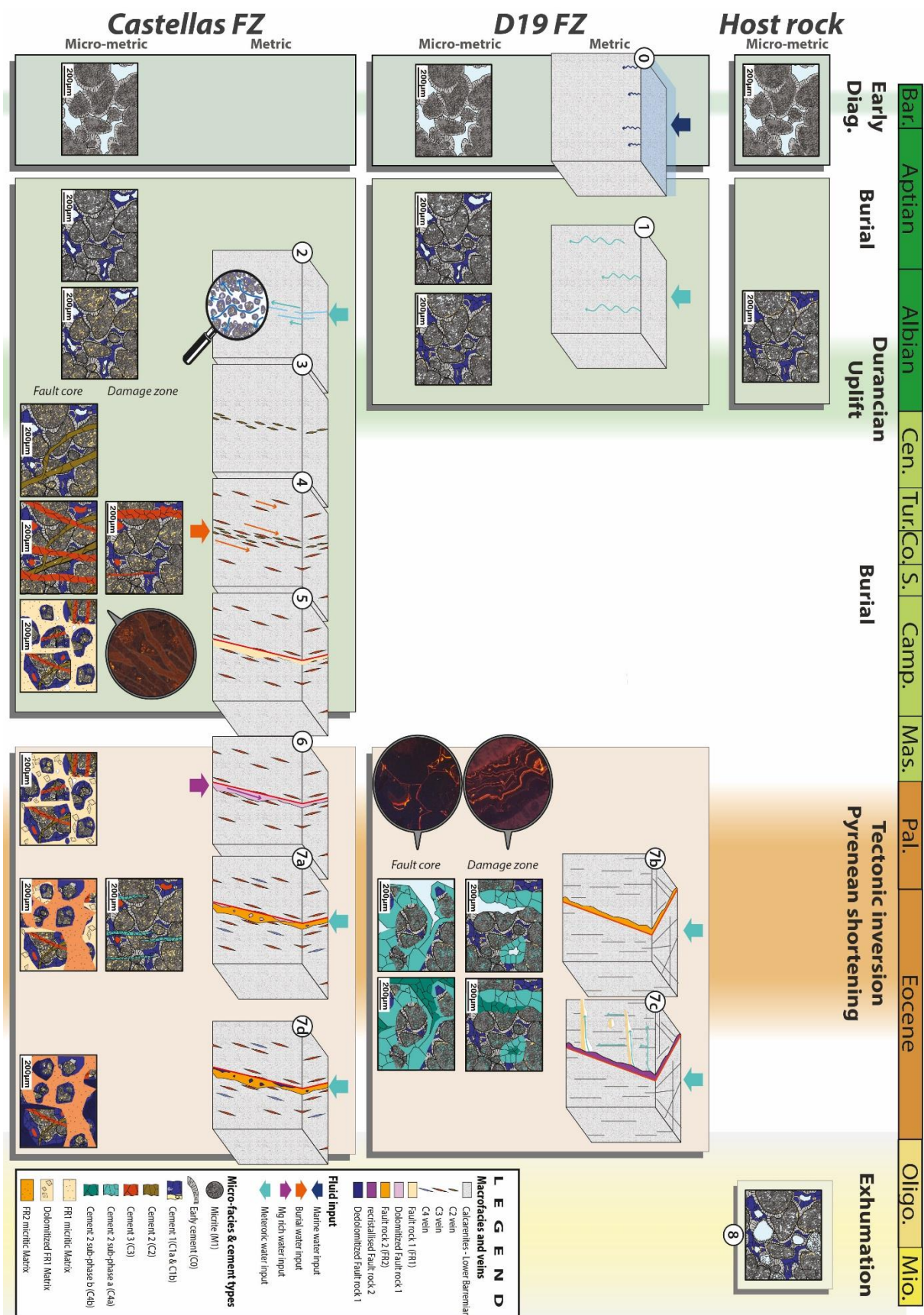


Figure 8 : Diagenetic and geodynamic evolution since the Barremian of both fault zones and host rock at the metric and micro-metric scale. Numbers 0 to 8 correspond to the steps 0 to 8 (see text for description).

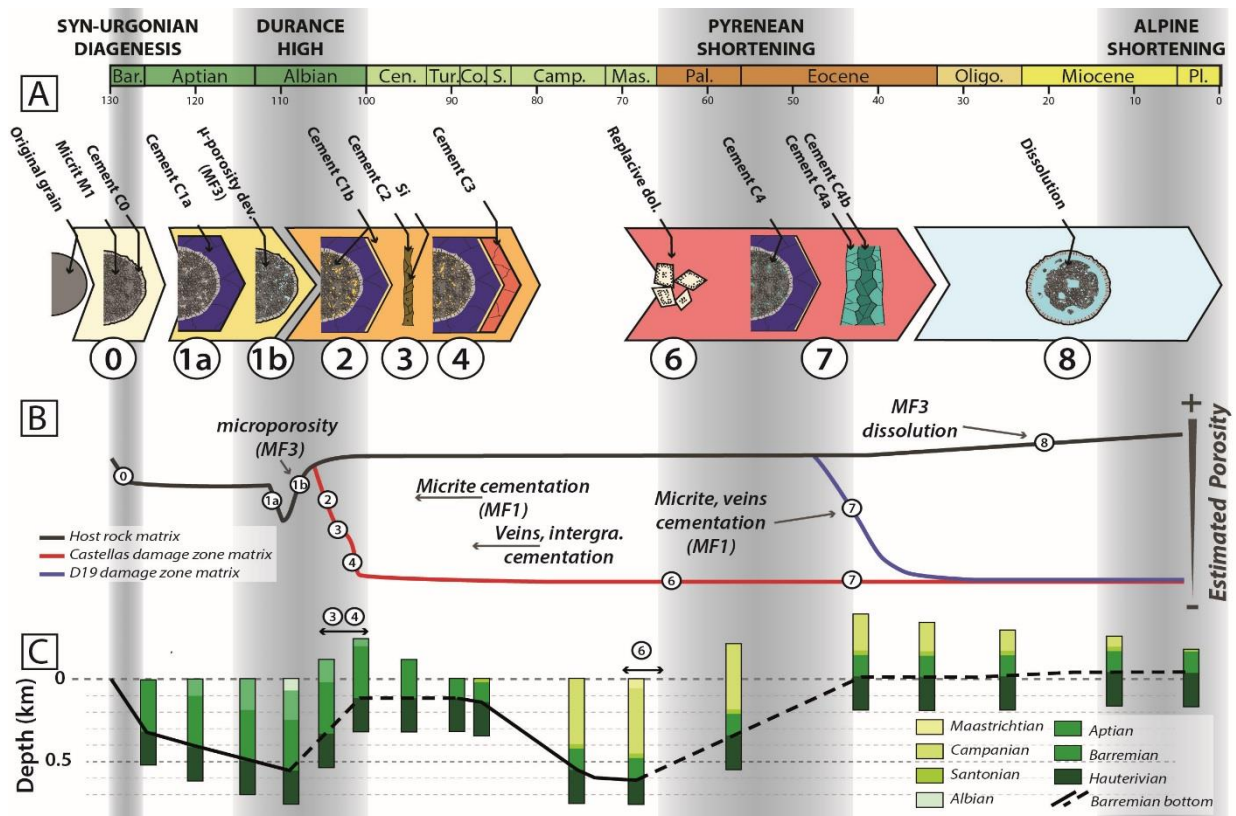


Figure 9 : Evolution of reservoir properties. A: different cementation phases; numbers 0 to 8 correspond to the steps 0 to 8 (see text for description), B: relative porosity evolution of the host rock and the 2 fault zones; C: Burial/Uplift curved of Barremian basement (modified from Matonti et al. (2012)).

Tectonic Inversion – Castellas fault-related dolomitization

At the onset of the Pyrenean shortening, compressive stresses lead to underground water upwelling through the permeable fault core. This fluid flow triggered the dolomitization of FR1 matrix (Step 6 on Fig. 8). This matrix-selective dolomitization can be favoured by several factors:

- (i) The matrix has higher permeability than cemented clasts with a smaller grain size, hence a higher grain surface area;
- (ii) This type of upwelling fluids, so-called “squeegee-type”, are short lived processes (Buschkuehle and Machel, 2002; Deming et al., 1990; Dorobek, 1989; Machel et al., 2000) not favourable for massive dolomitization;
- (iii) Low temperature fluids, under 50°-80°C, enabled the preservation of FR1 clast initial structure. Contrarily, high temperature dolomitization tends to be destructive (Machel, 2004);
- (iv) The tight surrounding host rock constrained high Mg fluid circulation to the fault core.

Gisquet et al. (2013) noticed similar fault related replacive dolomitization phase in the Etoile massif, 23km South-Est of the studied zones. They linked the dolomitization to compressive conditions during the early (Late Cretaceous) Pyrenean shortening. From these authors, the

tectonic stress led to low temperature upwelling fluids Mg-enriched by the dissolution of underlying Jurassic dolomites. The Jurassic dolomites also occur in La Fare anticline. Since the fluids leading to dolomitization of fault core were low temperature and since dolomites occur underground, it is possible that the dolomitization in La Fare and in the Etoile massif were similar and synchronous. Matrix dolomitization can increase inter-crystalline and/or inter-particle porosity up to 13% but the later dolomite overgrowth reduce the porosity and permeability (Lucia, 2004; Machel, 2004; Saller and Henderson, 2001). Hence, in the first stages of dolomitization, the fault core was an important drain. After the growth of dolomite crystals, the fault core turned to barrier (Fig. 9 B6 and C6)

Sinistral tectonic inversion – meteoric alteration of reservoir properties

The ongoing tectonic inversion with increasing compressive stresses finally led to the Castellás fault sinistral reactivation and to the onset of D19 fault zone (Aubert et al., 2019b). Aubert et al. (2019a) has shown that this compression reactivated the pre-existing early N030° background fractures (Step 7 on Fig. 8). This tectonic event lead to FR2 in fault cores but with specific diagenetic consequences. In the D19 fault zone, the fault nucleation and reactivation of back-ground fractures led to pluri-metric to kilometric fault surfaces with a permeable fault rock acting as drains and localizing the fluid flow (Aubert et al., 2019a). This fluid flow resulted in the cementation of C4a and C4b in veins and micritized grains (MF1, Step 7c on Fig. 8), what led to a strong porosity decrease in the fault zone (Fig. 9, B7 and C7). However, not all fractures were cemented by C4, so the fracture porosity/permeability was preserved. Therefore, the D19 fault zone became a type I reservoir sensu Nelson (2001) with a very low matrix porosity/permeability and high fracture permeability (Aubert et al., 2019a).

Along F2, successive fluids gave rise to karsts, karstic filling and dissolution/cementation of FR2 matrix (Step 7c on Fig. 8). Then, FR2 was sealed by C4 cementation. Isotopic values of C4 (from -9.2 to -6.1‰ for $\delta^{18}\text{O}$ and from -5.01‰ to -1.0‰ for $\delta^{13}\text{C}$) highlight the strong influence of meteoric fluids. This is coherent with the occurrence of karstic fill due to fluid circulations in vadose zone, alternating dissolution and cementation (Swart, 2015). However, the positive covariance between $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ of C4 suggests mixed fluids (Allan and Matthews, 1982) of meteoric water and burial or marine water.

In the Castellás fault zone, the host rocks are slightly impacted by these meteoric fluid circulations. Yet, some veins filled with C4a occur along transect 2 and transect 3 (Step 7a on Fig. 8). Two samples have higher $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ isotopic values (respective means of -6.25‰ and -4.20‰ for $\delta^{18}\text{O}$; -0.64 and -0.09‰ for $\delta^{13}\text{C}$) similar to C1 (Fig. 6A). This indicates that C4 in the Castellás fault zone was precocious in comparison to the D19. Cements C4 in Castellás area are restricted to transect 2. Transect 2 crosscut through the Castellás fault at the location of a relay zone (Fig. 2A). Relay or linkage zones occur where 2 fault segments overlap each other during fault grow (Kim et al., 2004; Long and Imber, 2011; Walsh et al., 1999, 2003). Consequently, the fault complexity, the fracture intensity and the fracture-strike range are increased (Kim et al., 2004; Sibson, 1996). This process in the studied area resulted in a well-connected fracture network that increased the local permeability and allowed local fluid circulations. In transect 2, the increase of the local permeability in the relay zone enhanced fluid flow related to cement C4. The relay zones along the Castellás fault and their consequences on the fracture permeability are, therefore, responsible for this local cementation event. Contrarily, cementation in D19 fault zone is linked to the highly permeable fault surfaces which acted as a drains (Aubert et al., 2019a). That implies that the cementation occurred only after the

formation of the fault surface. In the case of Castellás, the relay zone was already present, inherited from the former normal activity, allowing early C4 fluid to flow in fault zone. This, in addition, explains why the early C4 cementation has not been recorded in D19 fault zone. The C4 cementation in T2 reduced the porosity to less than 8% on a larger zone (>60m) than in both others transects (T1 ≈30m, T3>40m).

The reactivation of the Castellás fault formed a new fracture network that locally triggered the fracture connectivity and permeability. The Castellás fault zone formed a type I reservoir (Nelson, 2001), but lateral variation of the fracture network implies lateral variations of the hydraulic properties. Thus, the fault zone was both a drain and a barrier (Matonti et al., 2012), such as a sieve.

After these events, the matrix of the Castellás fault core was de-dolomitization (FR1) in relation to cementation C5 (Step 7d on Fig. 8). The C5 cement isotope values (mean of -7.49‰ for $\delta^{18}\text{O}$ and -4.01‰ for $\delta^{13}\text{C}$) are comprised within C4 positive covariance between $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$. This indicates a continuity between C4 and C5 fluid flows. The measurements with the SEM revealed a lack of Mg in the matrix indicating that C5 totally recrystallized the replacive dolomite. Following this de-dolomitization phase, no additional diagenetic event is recorded in Castellás fault zone.

A late Pyrenean to alpine compression reactivated the D19 fault zone what formed the new fault rock FR3. The matrix of this fault rock has very low $\delta^{13}\text{C}$ isotopic values (mean of -6.83‰) indicating an organic matter input (Swart, 2015). This implies soils, and thus results from a near surface fluid circulation. We deduce that the D19 faults was lately reactivated after the folding of the La Fare anticline. There is no such cementation with similar isotope values in the fault zone, meaning that fluids and cements did not alter the fault zone diagenetic properties.

Finally, the late exhumation of the Urgonian carbonate host rocks led to flows incurring dissolution of MF3 grains in the host rock. This phase triggered the moldic porosity and increased the porosity/permeability (Step 8 on Fig. 9B and C). These flows, however, did not affect fault zones.

2. EVOLUTION OF FAULT ZONES RESERVOIR PROPERTIES

The host rock presents a monophasic evolution and switch from a type IV reservoir where matrix provided storage and flow, to a type III reservoir where the fractures are pathways for flow but the production comes from the matrix (Nelson 2001, Fig. 10A). The fault zones present a more complex polyphase evolution than the host rock. Indeed, their reservoir properties evolved from a type IV reservoir corresponding to the host rock to a type I reservoir where fractures provide both storage and flow pathways (Nelson 2001, Fig. 10A). Both fault zones present slight differences. The Castellás fault zone was completely tight soon after C3 cementation. Consequently, it did not fit to the Nelson reservoir type classification. However, after fault core formation, the fault zone presents a high fault core permeability. In this study we propose a new approach with a triangle diagram taking into account fault core permeability to remove the flaws of this method (Fig. 10B). Thus, for Castellás fault zone, the permeability evolves from the host rock permeability (100% matrix; step 0 on Fig. 10B) to a permeability due to 50% to the matrix and 50% to the fault core during dilation band development (step 2 on Fig. 10B). Thereafter, during the 2 fracture events permeability is mainly link to fractures (C2: 30% FC, 70% fractures; C3: 15% FC, 15% matrix, 70% fractures; step 3, 4 on Fig. 10B). Then, after fault core formation and during dolomitization event, permeability is solely located in the

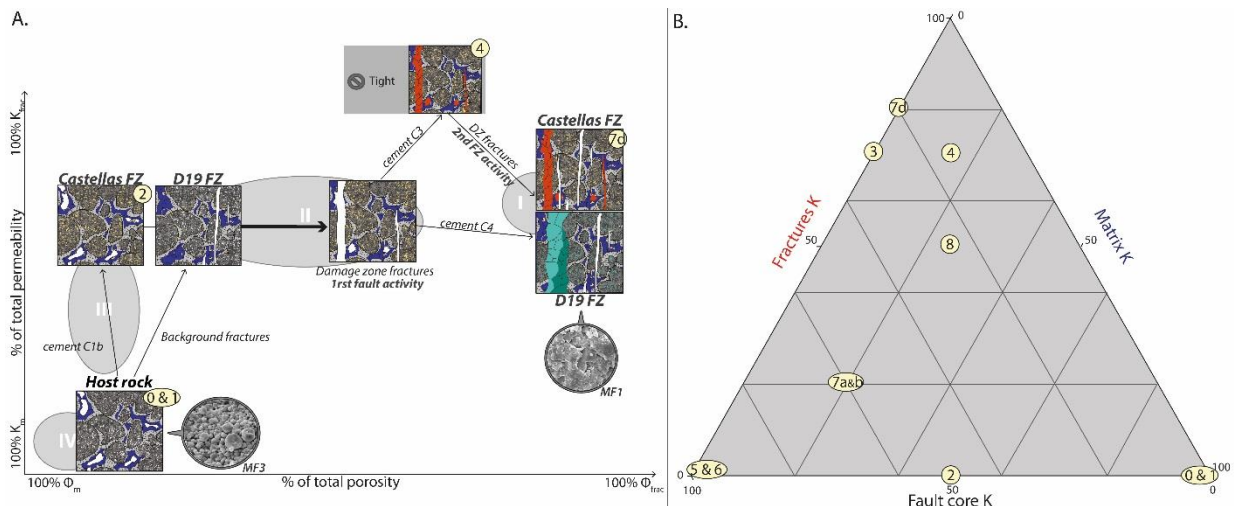


Figure 10 : Castellás and D19 fault zone reservoir properties evolution. A: evolution of permeability and porosity taking into account fault zone fractures and matrix after Nelson (2001) and B: Triangle diagram of permeability evolution with 3 components: matrix, fractures and fault core. Numbers 1 to 8 correspond to the steps 1 to 8 (see text for description).

fault core (step 6, 7 on Fig. 10B). Lastly, after fault zone reactivation, the permeability is due to 20% to the FC and 80% to fractures (step 7c on Fig. 10B). The D19 fault zone permeability during its development was related at 20% to the matrix, 20% to the fractures and 60% to the fault core (step 7a and 7b on Fig. 10B).

8. CONCLUSION

This study deciphered the diagenetic evolution of 2 fault zones and the impact on the reservoir properties of both fault and host rock in the frame of the overall geodynamic context of the SE basin. The main outcomes are:

- Fault zones may have a complex diagenetic history, but most diagenetic phases occur during the nucleation of the fault. In the case of Castellás fault zone, the diagenetic imprint is mainly influenced by early diagenesis occurring along fractures and diffuse dilation zones prior to the proper fault plane nucleation. Regarding D19 fault zone, most of diagenetic alterations occurred just after fault onset in the first stage of their activity. In both cases, the cementation altered initial reservoir properties in the fault zone vicinity, switching from type III to type I during the first stages of fault development. Later fault reactivation thinly impacts matrix porosity/permeability.
- Fault zones act as drains canalizing fluid flows in the beginning of their formation. This induces fault zone cementation but preservation of host rock microporosity. This important fluid drainage is visible on D19 outcrop where the flows led to dissolution/cementation of fault rock matrix and formed karsts.
- All diagenetic stages, including cementation and dolomitization, result from low temperature flows with important meteoric water input. This low temperature flows associated with the deformation and cementation types and, the lack of mineralisation specific to high temperature flows disprove any hydrothermal influence.

520 This regional study allows to draw broader rules for polyphase faults in granular carbonates at
521 low depth (Fig. 9).

- 522 • Under extensive context, fault nucleation can lead to dilation band acting as conduits
523 for fluid flow. Carbonates are very sensitive to fluid and rock-fluids interactions. Thus,
524 the onset of dilation bands triggers important diagenetic reactions that strongly alter
525 local reservoir properties. During later fault zone development, the diagenesis depends
526 on faults zones internal architecture.
- 527 • Fracture networks related to fault nucleation in granular carbonates form good fluid
528 pathways before proper fault plane formation. However, in the case of pre-fractured
529 carbonates, like D19 fault zone, fault rocks early appear in fault cores. In the later cases,
530 fluids flowed preferentially within the permeable breccia rather than the damage zone
531 fracture network.

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