Palaeoenvironmental context and significance of ferruginous tubular bioforms and other authigenic mineral formations in source-to-sink sedimentary systems

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Abstract

Surficial sediments on the seafloor from passive continental margins can provide insight into recent Late Quaternary sedimentary dynamics acting over offshore sedimentary systems. This work focuses on the study of some particular ferruginous tubular structures resembling bioforms (FTB) located in the distal Galician Continental Margin (NW Iberian Margin) at water depths between ~1550 and ~2200 m. The characterisation of these structures made it possible to study in depth their formation environment and subsequent sedimentary evolution during the Late Pleistocene and Holocene. The FTB consist of goethite with a framboidal texture. They were interpreted as formed by an initial pyrite precipitation in reducing micro-environments conditioned by the activity of sediment-dwelling organisms during the early diagenesis. This is followed by the oxidation of pyrite by a combination of hydrothermal fluids and erosional processes, which triggers the formation of the framboidal oxyhydroxides. The data allowed obtaining a comprehensive understanding of the environmental context and the significance of these ferruginous tubules, as there are no previous studies in the scientific literature that describe these structures in a source-to-sink sedimentary system.

Introduction

The study of the surficial sediments and geological structures of the seafloor is an essential tool for characterising the most recent sedimentary and oceanographic dynamics of continental margins. This is the case of the West Galician Continental Margin, located northwest of the Iberian Continental Margin, which has been the focus of several sedimentary infill studies, especially after the sinking of the Prestige oil tanker (e.g. Murillas et al. 1990; Ercilla et al. 2006, 2008, 2011). The study of the most surficial sedimentary cover of the margin revealed the palaeoclimatic and palaeoenvironmental interest of this area to understand in detail the climatic, environmental and oceanographic changes during the Late Pleistocene and Holocene at these latitudes (e.g. Alonso et al. 2008; Rey et al. 2008; Bender et al. 2012; Martins et al. 2013, 2015; Hanebuth et al. 2015; Plaza-Morlote

et al. 2017; Mena et al. 2018; Petrovic et al. 2019; López-Pérez et al. 2019, 2021a, b).

Despite the large number of works focused on the Galician Continental Margin, no previous work has focused on the attention and characterisation of the formation of ferruginous tubular bioforms (FTB). These materials were extracted and observed on the seafloor of the transitional zone (TZ) morphostructural province (Fig. 1), a geomorphological province defined by Vázquez et al. (2008), which corresponds to a general elevation of the marginal platform of the margin (López-Pérez et al. 2021a). The study is based on direct observations of the seabed and the geochemical and mineralogical analyses of these structures and other authigenic minerals from the surface, combined with the analysis of a sediment core for setting up the sedimentary dynamics of the study area.

In the scientific literature, there are very scarce examples of this type of structure in deeps marine environments (Rona et al. 1984; Thompson et al. 1985), so this study is a novelty in this field. However, ferruginous structures of different types have been described in the fossil record as in the Middle Jurassic condensed sequences and hardgrounds in the Southern Carpathians (e.g. Lazăr et al. 2013; Lazăr and Grădinaru 2014). A better understanding of the current

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Fig. 1 Multibeam bathymetry map of the Galician Continental Margin with the locations of the six main morphostructural provinces: Galicia Interior Basin (GIB), Transitional Zone (TZ), Galicia Bank (GB), Northwestern Flank (NF), Deep Galicia Margin (DGM) and Half-Graben Domain (HGD) morphostructural provinces. The red circle indicates the position of core GC16 ($42^{\circ} 41' 37.70''/11^{\circ} 02' 31.10''$; length 23 cm; water depth 1734 m), and green and blue lines refer to the locations of the Agassiz trawl and the ROV observation tracks, respectively. The multibeam data were obtained using a multibeam echosounder during the MARBANGA, Gran Burato 4240 and GALINCLIMARCH cruises, combined with the bathym-

etric survey data from the EMODnet Digital Terrain Model repository (emodnet-bathymetry.eu) (modified from López-Pérez et al. 2021a). The location map also includes a schematic representation of the current circulation of the superficial, intermediate and deep water masses around the West Iberian Continental Margin (modified from Hernández-Molina et al. 2011; Llave et al. 2019). The water masses are ENACW_{sp}=Eastern North Atlantic Central Water (subpolar); ENACW_{st}=Eastern North Atlantic Central Water (subtropical); MOW=Mediterranean Outflow Water; LSW=Labrador Sea Water; NADW=North Atlantic Deep Water; and LDW=Lower Deep Water

genesis of the ferruginous tubular bioforms will allow the scientific community to interpret ancient sedimentary environments in more detail. Therefore, this work enabled obtaining key information about the palaeoenvironmental context of the formation of these singular structures and their subsequent erosion from the Late Pleistocene to the present.

Study area

Location and physiography of the study area

The West Galician Continental Margin is a passive rifted margin characterised as a non-volcanic and hyperextended

continental margin (Boillot et al. 1979; Tucholke et al. 2007). The margin of Galicia has been divided into six morphostructural provinces (Murillas et al. 1990; Reston 2005; Vázquez et al. 2008), named from the continental shelf to the west as the Galicia Interior Basin, Transitional Zone, Galicia Bank, Northwestern Flank, Deep Galicia Margin and Half-Graben Domain (Fig. 1). This physiography is the result of a complex tectonic evolution caused by the Mesozoic rifting stages and the Cenozoic compressional regimes (Pyrenean orogeny) (Vázquez et al. 2008).

The study area is located at the Transitional Zone (TZ) morphostructural province (see coordinates in Fig. 1), and it corresponds to an elevated and isolated marginal platform located in a distal zone of the Galician Continental Margin (Vázquez et al. 2008; López-Pérez et al. 2021a). This province is defined as an NW–SE elongated province located between water depths of 1600 and 2500 m with seafloor gradients between $<0.5^\circ$ and 3° (Ercilla et al. 2011; Somoza et al. 2019). The high marginal platform is also characterised by a set of several structural highs and scarps and by a general topographic uplift in the centre, resulting in a dome-like morphology that creates an irregular-subrounded sloping seafloor (Ercilla et al. 2011; Somoza et al. 2019; López-Pérez et al. 2021a). Also, three giant depressions related to fluid seepages have been described, the largest of them named *Gran Burato* or *Burato ERGAP* (Vázquez et al. 2009; Ribeiro 2011; Ercilla et al. 2011; Druet 2015; Somoza et al. 2019; Minshull et al. 2020; López-Pérez et al. 2021a). These structures are interpreted as gravitational collapses due to large-scale fluid migration related to diapirism. Their origin could be related to the mobility of the “black shale unit” of Albian–Cenomanian age, which was identified as the possible agent responsible for the intense faulting activity in the study area (Group Galice 1979; Vázquez et al. 2008, 2009; Ercilla et al. 2011; Somoza et al. 2019; López-Pérez et al. 2021a). Furthermore, the Late Quaternary sedimentary evolution of this high marginal platform is strongly controlled by neotectonics and by the intensity of the hugging-bottom currents that erode the surficial sediment (López-Pérez et al. 2021a).

Occurrence of fluid activity in the study area

Evidence of hydrates and shallow gas on the marginal platform is unclear. Several seafloor expressions associated with fluid scapes (pockmarks) have been identified in the study area with a wide range of sizes and depths (Vázquez et al. 2009; Ribeiro 2011; Ercilla et al. 2011; Druet 2015; Somoza et al. 2019; Minshull et al. 2020; López-Pérez et al. 2021a).

Stratigraphic analyses display evidence of fluid migrations and accumulations in deep and shallow stratigraphic units in the *Gran Burato* pockmark area, the largest depression of the study area. This depression also shows a strong

structural control, so faults and fractures acted as paths during the fluid seepage responsible for its formation (Vázquez et al. 2009; Ribeiro 2011). Furthermore, some seismic blanking zones have been identified in the *Gran Burato* area. This fact was interpreted as an evidence of the presence of gas in the sediments (Vázquez et al. 2009; Ribeiro 2011; Minshull et al. 2020). On the other hand, the polarity inversion associated with the hydrate bottom simulating reflector (BSR), a direct indicator for hydrates, have not been identified in the TZ morphostructural province (Minshull et al. 2020). The analyses of the sediments did not report direct indicators of hydrates such as thermal anomalies, chlorinity anomalies or significant sulphate depletions (Minshull et al. 2020). Finally, benthonic fauna related to gas seepage, but not exclusive to these environments, was identified in the study area (Minshull et al. 2020).

Oceanographic settings

The study area is under the influence of several water masses, the most important of which is the Eastern North Atlantic Central Water that reaches depths of around 650 m, the Mediterranean Outflow Water that extends down to a depth of 1750 m and the Labrador Sea Water that extends down to depths of 1500 to 2200 m (Fiúza et al. 1998; Hall and McCave 2000; McCave and Hall 2002; Varela et al. 2005; Zhang et al. 2016). Seafloor depths of less than 3000 m in the TZ and the GIB provinces constitute a geomorphological barrier for deeper water masses such as the North Atlantic Deep Water (2150–3450 m water depth) and the Lower Deep Water (>3450 m water depth) (Bender et al. 2012).

Methodology

ROV observations and sediment sampling

Direct observations of the deep ocean floor were obtained using a *Seaeye Cougar 1432* ROV (remotely operated vehicle) from the *ACSM company* during the *Burato 2011* cruise. A total of 45 h of video recording of six ROV dives were obtained (Fig. 1).

The sediment samples from the seafloor were collected from the surface using an Agassiz trawl during the *Gran Burato 2011* cruise (Fig. 1). The trawl mouth was 3.5 m wide and 0.7 m vertical opening, and the net mesh size was 10 mm. Also, a short sediment core (23 cm length), labelled as GC16 and extracted during the *MARBANGA* cruise, was studied (Fig. 1).

Geochemical and sediment analyses

High-resolution X-ray fluorescence (XRF) and magnetic susceptibility (MS) profiles of the core, as well as

its optical and radiographic images, were obtained with the Itrax™ Core Scanner at the research support services (CACTI) of the University of Vigo. XRF data were acquired with a Mo-tube operated at a voltage of 30 kV and an exposure time of 20 s. Semi-quantitative XRF raw data were obtained every 1 mm and smoothed using a 10-point running mean (1 cm) following the recommendations of Rodríguez-Germade et al. (2013). Grey levels (GL) and RGB values were obtained from radiographic and optical images, respectively.

Grain-size distributions were determined from discrete samples collected every 2 cm using a laser diffraction particle size analyser, the Coulter LS 13 320 (Beckman Coulter Inc., Brea, CA) from the University of Vigo.

The mineralogy of the core and sediment samples was studied by X-ray diffraction (XRD), using an X'Pert Pro (PANalytical) diffractometer, and by the JEOL JSM-6700f scanning electron microscope (SEM) operated in backscattering mode (BS) at the University of Vigo. Moreover, FTB were explored by transmission electron microscopy (TEM) using a JEOL JEM-2010 FEG (200kV) microscope and by scanning electron microscopy using a Dual-Beam FEI Helios Nanolab microscope at the CACTI of the University of Vigo.

Carbon and oxygen stable isotope analyses were performed in calcite and dolomite samples to characterise their possible methanogenic origin. The measurements were carried using a GasBench II system coupled to a *DELTA V Advantage* isotope ratio mass spectrometer (IRMS) from the Autonomous University of Madrid. The isotope data are reported as per mil values in δ notation relative to the international Vienna Pee Dee Belemnite (VPDB) standard.

Stable Sr isotope ratios ($^{87}\text{Sr}/^{86}\text{Sr}$) of FTB and carbonate and phosphate samples collected using an Agassiz trawl were measured using a Thermo-Finnigan (Neptune) Multi-collector inductively coupled plasma mass spectrometer (MC-ICP-MS) according to the internal protocol of the CACTI in the University of Vigo. Finally, isotopic compositions of trace and rare earth elements of FTB were obtained by inductively coupled plasma-mass spectrometry (ICP-MS) in an X Series ICP-MS (Thermo Elemental) at the CACTI of the University of Vigo.

Multivariate statistical analyses

Statistical facies analyses of the core samples were carried out using the SPSS v.23 software using the procedure described in López-Pérez et al. (2019). The facies classification was based on the application of several multivariate statistical analyses (cluster analysis and discriminant analysis) to the large high-resolution geochemical, magnetic and sedimentological dataset from the core.

Core chronology

The chronology of the core GC16 was previously reported in López-Pérez et al. (2021a). This age model was obtained from four monospecific handpicked samples of the planktonic foraminifera *Globigerina bulloides*. These samples were analysed for radiocarbon dating by accelerator mass spectrometry (AMS) (Beta Analytic, Miami, Florida, USA). The ^{14}C ages were converted to calendar ages using the IntCal13 calibration curve (Reimer et al. 2013), including a constant of 400-year reservoir correction (Plaza-Morlote et al. 2017).

The age model was developed using a stratigraphy-based Bayesian approach running the OxCal v4.3.2 software (Ramsey 2008, 2017). The 100% overall agreement calculated confirms statistically the robust model obtained.

Results

ROV observations

Direct images from the seafloor allowed identifying FTB structures distributed over flat surfaces, along with accumulations of pteropods and gravels (Fig. 2a, b). FTB have also been identified within rock fragment trails associated with comet marks that are aligned with the direction of the current behind the dropstones (Fig. 2c).

Furthermore, the ROV observations allowed the identification of dense carbonate-like crusts with laminated morphology and partially covered by sand (Fig. 2d, e, f).

Characterisation of the surficial sedimentary materials

Different typologies of sedimentary materials were collected from the seafloor of the *Gran Burato* surrounding area in the TZ morphostructural province using an Agassiz trawl (Fig. 1). These sedimentary materials were identified and classified into three categories: ferruginous tubular bioforms (FTB) and carbonate and phosphate sediment samples (Figs. 3, 4).

Ferruginous tubular bioforms

The FTB collected from the seafloor (also observed directly with the ROV, Fig. 2) are characterised by presenting a tubular morphology, some of which have dendritic or arborescent ramifications (Fig. 3a). These FTB have a mean length of 6.79 cm, with minima and maxima of 3 and 15.50 cm, respectively, and a mean diameter value of 0.71 cm, with minima and maxima of 0.40 and 1.70 cm, respectively. More

Fig. 2 Seafloor observations of the study area obtained with the ROV: (a) accumulation of ferruginous tubular bioforms (FTB) and pteropods on a flat surface; (b) detailed photograph of the accumulation

rarely, these samples show an irregular tabular morphology with an average length, width and thickness of 5.82, 1.68 and 0.90 cm, respectively (Fig. 3b). Some of the tubules

of ferruginous tubular bioforms (FTB) and pteropods on the seafloor; (c) comet mark with an accumulation of FTB; (d, e and f) images of dense crusts

are hollow and have concentric growth structures (Fig. 3c). Generally, the terminations of the tubules have a fractured appearance.

Fig. 3 Photographs of the ferruginous tubular bioforms (FTB) collected from the seafloor by the Agassiz trawl (BV-1 and BV-2; see location in Fig. 1): (a) ferruginous tubular bioforms; note that some samples have irregular tabular or arborescent morphologies; (b) FTB with irregular tabular morphologies; (c) detail of a hole (left) and a concentric growth structure (right) in an FTB; (d) SEM image with the superposition of the elemental mapping showing Fe in green and O in red, accompanied by an XRD diffractogram of a goethite sample

Fig. 4 Photographs of (a) and (b) calcite; (c) dolomite; (d) and phosphate samples collected from the seafloor by the Agassiz trawl (BV-1 and BV-2; see location in Fig. 1)

XRD, SEM and TEM analyses show that goethite is the unique constituent of these FTB (Figs. 3d, 5). Moreover, SEM and TEM observations allowed to differentiate framboidal structures of goethite within FTB (Fig. 5). These framboids were characterised as polycrystalline structures composed of

nano-sized crystals (Fig. 5d, f). Surrounding the framboids, there are more massive goethite crystals with slightly larger crystal size without framboidal texture (Fig. 5e).

The stable Sr isotope ratios ($^{87}\text{Sr}/^{86}\text{Sr}$) of seven ferruginous structures display values between 0.70950 ± 0.000033

Fig. 5 (a and b) SEM images obtained by the Dual-Beam FEI Helios Nanolab microscope of the framboidal structures of goethite; (c) TEM image of the framboidal texture within FTB; (d) detailed image of the nano-sized crystals that compose the polycrystalline framboids; (e) TEM image of the framboidal structures and the massive goethite crystals; (f) diffractogram of polycrystalline material measured in the FTB sample corresponding to goethite

and 0.71005 ± 0.000043 (Fig. 6a). Furthermore, Table 1 shows the concentrations of the trace and rare earth elements measured in the FTB. These data were normalised using the North America Shale Composite (NASC) reference values published by Gromet et al. (1984) (Fig. 6b). The results show that FTB are enriched in radiogenic elements such as strontium and uranium (Fig. 6b). The absence of the negative Ce anomaly in seawater is also noteworthy. Furthermore, in Fig. 6b, it can be seen that

heavy and light elements present similar normalised values.

Carbonate samples

XRD analyses and SEM observations of the carbonate fragments collected with the Agassiz trawl allowed to differentiate between calcite and dolomite samples (Fig. 4a, b, c). Calcite samples show a sub-rounded morphology

Fig. 6 **a** Stable Sr isotope ratios ($^{87}\text{Sr}/^{86}\text{Sr}$) measured in five samples of FTB, calcite, dolomite and fluorapatite collected from the seabed using an Agassiz trawl. The red line indicates the modern seawater composition of $^{87}\text{Sr}/^{86}\text{Sr}$. The high $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic ratios obtained from FTB in comparison to the modern seawater have led to a consideration of hydrothermal activity in the study area; **(b)** ternary diagram of trace elements (Sr, Rb and Ba), and NASC-normalised data of the trace and rare earth elements measured in FTB

Table 1 Isotopic compositions of the trace and rare earth elements of seven samples of FTB

Isotope	T1 (mg/kg)	T2 (mg/kg)	T3 (mg/kg)	T4 (mg/kg)	T5 (mg/kg)	T6 (mg/kg)	T7 (mg/kg)
^{85}Rb	2.474	5.411	3.946	3.644	3.476	5.366	4.416
^{88}Sr	64.370	66.890	66.400	55.800	67.360	68.420	77.960
^{133}Cs	0.273	0.632	0.537	0.362	0.405	0.554	0.445
^{137}Ba	13.420	22.360	12.770	12.250	14.430	17.170	17.190
^{139}La	1.270	1.140	1.386	1.145	1.277	1.102	1.431
^{140}Ce	1.666	2.104	1.470	1.429	2.528	1.833	1.970
^{146}Nd	1.217	1.080	1.189	0.893	1.172	0.956	1.245
^{147}Sm	0.292	0.225	0.253	0.186	0.262	0.205	0.269
^{153}Eu	0.075	0.054	0.061	0.045	0.068	0.048	0.062
^{159}Tb	0.061	0.035	0.040	0.032	0.049	0.033	0.045
^{172}Yb	0.311	0.131	0.158	0.123	0.188	0.119	0.168
^{175}Lu	0.049	0.020	0.026	0.019	0.030	0.020	0.028
^{232}Th	0.381	0.324	0.483	0.517	0.427	0.291	0.426
^{238}U	2.596	1.138	0.970	0.740	2.771	1.086	1.361

(Fig. 4a, b) and residual amounts of ankerite (2%) detectable by XRD in some of them. The samples have an average length, width and thickness of 2.74, 1.80 and 0.93 cm, respectively. Dolomite samples show a planar morphology with an average length, width and thickness of 5.84, 4.58 and 0.50 cm, respectively (Fig. 4c).

The $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic ratios display values between of 0.70829 ± 0.000017 and 0.70904 ± 0.000066 in three calcite samples, and values of 0.70828 ± 0.000070 and 0.70874 ± 0.000037 in two dolomite samples (Fig. 6a). The $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ isotopic compositions in calcite show values of $\delta^{13}\text{C} = -0.32\text{‰}$, and $\delta^{18}\text{O} = -5.13\text{‰}$; meanwhile, the dolomite sample displays values of $\delta^{13}\text{C} = 0.20\text{‰}$, and $\delta^{18}\text{O} = -3.58\text{‰}$.

Phosphate samples

XRD analyses showed fluorapatite and hydroxyfluorapatite as the main mineralogical constituents of two of the samples obtained with the Agassiz trawl (Fig. 4d). These samples have a rounded morphology, with an average length, width and thickness of 6, 3.50 and 3.50 cm, respectively.

The $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic ratios measured in two samples are 0.70926 ± 0.000028 and 0.70952 ± 0.000033 (Fig. 6a).

Sediment core data

To interpret the extension and formation of these authigenic minerals, the core GC16, close to the study area, was also studied. This core displays a homogeneous distribution of the sand (44.01%), silt (36.94%) and clay (19.05%) fractions

throughout the core (Fig. 7). The mean grain size (MGS) and sorting show a mean value of $97.30 \mu\text{m}$ ($\phi = 3.36$) and $106.76 \mu\text{m}$ ($\phi = 3.23$), respectively. Thereby, the sedimentary texture of this core is defined as very poorly sorted, very fine sand. This description is in agreement with other sandy cores studied in the study area (López-Pérez et al. 2021a, b).

Furthermore, the core GC16 exhibits two very different facies: a *low-density sand* (*Low-den sand*) facies (Figs. 7, 8a, b) at the top of the core and a *high-density silt* (*High-den silt*) facies (Figs. 7, 8c, d) at the bottom of the core. The boundaries of these facies coincide with changes in RGB colour and GL profiles (Fig. 7). This facies classification was carried out using multivariate statistical analyses applied to geochemical (XRF, Fe, Ti, Ca, Mn; and Ba, XRF Fe/Ca, Ti/Ca, and Si/Sr detrital ratios), sedimentological (grain-size, grey level, and RGB colour), and magnetic (MS) data (Fig. 7) (López-Pérez et al. 2021a), using the methodology applied in López-Pérez et al. (2019).

Low-density sand facies

This facies comprises well-preserved foraminifera accompanied by detrital carbonates and siliciclastic fragments of different sizes in a well-sorted coccolithophoridae matrix (Fig. 8a, b). Moreover, SEM observations display sparse iron oxides.

The facies shows an average XRF-Ca value of $111,222.63 \pm 11,540.16$ p.a. and mean values of XRF-Fe, Ti and MS of $10,900.76 \pm 1150.81$ p.a., 231.97 ± 37.38 p.a. and $7.15 \pm 4.81 \cdot 10^{-5}$ SI for each proxy, respectively (Fig. 7). The XRD analyses indicate that this facies is mainly composed of 90% calcite, 6% magnesian calcite and 4% quartz.

Fig. 7 Facies classification obtained from the core GC16. Depth plot of the facies description followed by an optical and radiographic image, grain size distribution, magnetic susceptibility, XRF-Fe/Ca, Ti/Ca, and Si/Sr detrital ratios, XRF-Ca, RGB values and grey level (GL) of each core. Note the change in colour and the increase in density and content of the fine fraction in the high-density silt facies. The grey rectangle indicates a deteriorated core level

Fig. 8 SEM micrographs obtained at different depths illustrating the described facies of core GC16: (a) low-density sand facies at 8 cm in core GC16 composed of well-preserved foraminifera, accompanied by detrital carbonates and siliciclastic fragments of different sizes, in a well-sorted coccolithophoridae matrix; (b) magnified optical image of this level where it is possible to recognise the coccolithophoridae matrix; (c) high-density silt facies at 19 cm in core GC16, also comprised by of well-preserved foraminifera with detrital carbonates and siliciclastic fragments of different sizes, in a well-sorted coccolithophoridae matrix; (d) magnified optical image of this level where it is possible to recognise the coccolithophoridae matrix

High-density silt facies

This horizon consists of well-preserved foraminifera sands within a coccolithophoridae matrix. SEM analyses show the presence of terrigenous components such as detrital carbonates and siliciclastic fragments of different sizes, as well as iron and titanium oxides with high backscatter coefficient (Fig. 8c, d). All these detrital components are more abundant than in the previous *Low-den sand* facies.

The *High-den silt* facies exhibits a mean XRF-Ca content of $128,211.04 \text{ p.a.} \pm 10,022.22 \text{ p.a.}$ and average values of XRF-Fe, Ti and MS of $7523.38 \pm 2777.70 \text{ p.a.}$, $193.84 \pm 96.48 \text{ p.a.}$ and $7.23 \pm 17.30 \cdot 10^{-5} \text{ SI}$, respectively (Fig. 7). Regarding XRD analyses, the facies is composed of 88% calcite, 8% magnesian calcite and 4% quartz.

This facies shows textural, geochemical and magnetic properties similar to those of *Low-den sand* facies. However, *High-den silt* facies shows a very coarse silt texture ($51.86 \mu\text{m} - \phi = 4.27$) and a higher density ($GL = 33,506.61$), in comparison with the previous one, which exhibits a very fine sand texture ($121.53 \mu\text{m} - \phi = 3.04$) and lower density ($GL = 33,706.40$) (Fig. 7). The main difference between them is their different colours. Whilst the *High-den silt* facies shows a white colour (2.5Y8/1), the *Low-den sand* facies displays a light yellowish brown (2.5Y6/4) tonality

(Fig. 7). In addition, no goethite tubules were found in this core, and the carbon and oxygen isotope data of both facies, reported by López-Pérez et al. (2021a), reflect the isotopic conditions of marine carbonates.

Sedimentation rate values

Core GC16 displayed very low sedimentation rates during the last 30 cal ka BP (mean of 1.57 cm ky^{-1}) with a hiatus between 17.80 and 10.45 cal ka BP, coincident with the abrupt colour change and the boundary between facies (Fig. 9).

Discussion

Sedimentological significance of ferruginous tubular bioforms

The ferruginous tubules of goethite described have been interpreted as bioform-like structures distributed over the surficial sediments of the study area. A temporal sequence based on burrowing-precipitation-venting-erosion-distribution processes has been described to explain the palaeoenvironmental context and significance of the FTB (Fig. 10).

Fig. 9 Depth plot with the facies description of core GC16, accompanied by the optical image and the age model obtained using the OxCal v. 4.3.2 software. Note the hiatus calculated between 17.80 and 10.45 cal ka BP, coincident with an abrupt colour change between the two facies described for this core. R_dates are the calibrated ages obtained from the radiocarbon dates (^{14}C)

Diagenetic precipitation of the FTB in sediments

Firstly, macrozoobenthos activity creates vertical and slightly branched burrows within the sedimentary record (Fig. 10a). Inside these burrows, iron precipitation triggers the FTB formation (Fig. 10a). Benthic fauna studies allowed identifying sediment-dwelling organisms in the study area, such as *Sipuncula* or *Polychaeta* (Rey and Gran Burato Science Team 2011). The presence of these species supports the hypothesis of the FTB formation inside the burrows.

The SEM analyses show polycrystalline framboidal textures composed of nano-sized goethite crystals within the FTB (Fig. 5). In the scientific literature, few articles describe framboidal oxyhydroxides in marine environments, and these studies are located mainly in the Gulf of Cadiz (Merinero et al. 2008, 2009; González et al. 2009, 2010). Merinero et al. (2008) recognised framboidal goethite in hydrocarbon seep-related carbonate chimneys in the Gulf

of Cadiz; meanwhile, González et al. (2009) identified framboids of goethite in Fe–Mn nodules in the same study area. These authors interpreted the formation of these framboidal oxyhydroxides as a result of pseudomorphism after pyrite.

For these reasons, FTB could have originally been formed by pyrite precipitation in reducing microenvironments by the activity of organisms within burrows during the early diagenesis. In addition, the presence of holes and concentric growth structures in some of the FTB (Fig. 3d, e) suggests a concentric precipitation growth of the tubules from the wall to the inside of the burrows, confirming their diagenetic origin.

Oxidation of the framboidal pyrite

After that, a combination of hydrothermal activity and erosional processes could provoke the oxidation of pyrite (Fig. 10b). The alteration of the framboidal pyrite could

Fig. 10 Schematic representation of the formation of the FTB: (a) burrows created by macrozoobenthos and pyrite genesis (P) during early diagenesis; (b) oxidation of pyrite to goethite (G) by hydrothermal activity and by the action of bottom currents, preserving the framboidal texture; (c) exhumation, remobilisation and horizontal redistribution of the FTB over the seabed due to the activity of a high-velocity bottom-current

explain the framboidal texture of goethite within the FTB. This process allows preserving the diagenetic signal of the formation of the FTB.

Hydrothermal activity in the study area is demonstrated by the $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic ratios. The high values of stable Sr isotope ratios ($^{87}\text{Sr}/^{86}\text{Sr}$) measured in the

ferruginous tubules (values between 0.70950 ± 0.000033 and 0.71005 ± 0.000043) are higher than in modern seawater ($^{87}\text{Sr}/^{86}\text{Sr} = 0.709190\text{--}0.709200$) (González et al. 2012). This suggests a source of radiogenic Sr, which can only be explained by the effect of diffuse hydrothermal venting in the study area (Fig. 6a), as suggested by González et al. (2012) in ferromanganese nodules in the Gulf of Cadiz. Likewise, the enrichment in Sr and U of the FTB, and the absence of the negative anomaly of Ce in seawater (Fig. 6b), could confirm the hydrothermal activity. Furthermore, the enrichment in heavy elements in relation to light elements (Fig. 6b) supports the hydrothermal activity hypothesis.

This hypothesis also agrees with the tectono-sedimentary context of the high marginal platform. In this study area, previous studies have been described faults that can act as conduits for the escape of fluids (Vázquez et al. 2009; Ribeiro 2011; López-Pérez et al. 2021a). This fact makes it possible to explain the context in which hydrothermal activity occurs.

Previous studies also demonstrated continuous oxygenation of the sediment of the study area (López-Pérez et al. 2021b). This fact could contribute to the oxidation of the pyrite by the action of oxygen-rich bottom waters.

Exhumation of the FTB

Afterwards, these FTB structures were exhumed, remobilised and redistributed over the seabed by high-velocity bottom currents (Fig. 10c) as displayed in ROV images (Fig. 2a, b, c). The direct FTB observations distributed over flat sea-floor surfaces and comet marks suggest the presence of high-velocity bottom-current acting in the study area. Other sedimentary structures identified by López-Pérez et al. (2021a), such as grooves and ripples, confirms erosional activity by bottom-hugging currents in the high marginal platform. The erosive and remobilising activity of a bottom-hugging current could explain the current horizontal and exhumed distribution of the FTB structures over the seafloor of the study area, as well as the fractured appearance of most of them (Fig. 10c).

Therefore, the FTB accumulation over the deep ocean floor could be an indicator of a present-day vigorous bottom-hugging current acting and eroding the sediments of deep-marine environments, such as the sediments of the transitional zone morphostructural province. This hypothesis is also supported by the extremely low sedimentation rates obtained from the sediment core (Fig. 9) and from other sediment cores of the study area (López-Pérez et al. 2021a). These low values contrast with those obtained in previous studies in other provinces of the Galician Continental Margin. Rey et al. (2008) reported sedimentation rate values around 9.1 cm/ka in MIS 2 and 3 cm/ka during the Holocene at the Half-Graben Domain morphostructural province. Regarding the Galicia Interior Basin, Martins et al. (2013)

reported values between 2.4 and 7.7 cm/ka during MIS 2 and MIS 3, and between 2.7 and 4.5 cm/ka during the Holocene; meanwhile, Mena et al. (2018) calculated sedimentation rates around 10 cm/ka east of the Galicia Interior Basin and 5 cm/ka west of this province before the Last Glacial Maximum (LGM), reaching maximums of 30 cm/ka during the LGM and Heinrich Events. On the contrary, this study reported values around 2 and 3 cm/ka for the Holocene at the Galicia Interior Basin. This contrast between sedimentation rates between morphostructural provinces supports the presence of bottom currents with high erosive capacity acting at the high marginal platform. These environmental conditions favour the erosion and exhumation of the FTB in the study area.

Sedimentological significance of the facies classification for the formation and distribution of the FTB

Core GC16 presented two facies, the *Low-den sand* facies and the *High-den silt* facies (Figs. 7, 8, 9). The boundary between them coincides with an abrupt change in colour and density. Moreover, this boundary is also coincident with a hiatus period, ranging from 17.80 to 10.45 cal ka BP (Fig. 9). This hiatus coincides with the time range where an increase in the strength of the bottom current is documented in the Galician Continental Margin related to the strengthening of the Mediterranean Outflow Water (MOW) (Petrovic et al. 2019). Therefore, the hiatus of the core GC16 indicates an increase in the strength of the bottom current that causes erosion of the sediments (López-Pérez et al. 2021a), and thus the exhumation of the FTB.

The sedimentological results of the core are also coincident with other sandy cores studied in the study area (López-Pérez et al. 2021a, b). These sediments with a high content of sand fraction, where the presence of benthic fauna has been described (Rey and Gran Burato Science Team 2011), generate the environment for the formation of FTB in abandoned burrows.

Additionally, the $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values related to marine carbonates for both facies of the core GC16 (López-Pérez et al. 2021a) suggest that the high-density facies is not related to methane-derived authigenic carbonates associated with recent escapes of methane-rich fluids. This fact confirms that there is no recent evidence of methanogenic activity in the study area during the Late Quaternary.

Interpretation of the authigenic carbonates and phosphates

Furthermore, the recognition of authigenic minerals such as apatite, calcite and dolomite fragments over the seafloor

of the study area (Fig. 4) also constitutes an indication of the erosional activity due to the action of a bottom-current.

These authigenic minerals precipitated within the sediment column and subsequently were eroded and remobilised by the bottom-current activity. The low $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic ratios obtained for the carbonate samples in comparison to the FTB suggest a non-radiogenic origin (Fig. 6a). In the case of calcite and dolomite fragments, the low $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic ratios obtained with regard to the modern isotopic values of seawater (Fig. 6a) could suggest an isotopic exchange between carbonates and seawater when the samples were exposed to erosional activity on the seafloor (González et al. 2012). Besides, the $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ isotopic values obtained from calcite and dolomite samples reflect the isotopic conditions of marine carbonates (Joseph et al. 2013), indicating that these samples are not related to methane-derived authigenic carbonates associated with methane migration. These carbonate materials could be fragments of the dense carbonate-like crusts observed from the ROV (López-Pérez et al. 2021a). Regarding the apatite samples, the $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic ratio of sample number 13, coincident with the isotopic values of modern seawater (Fig. 6a), indicates a non-radiogenic origin and non-isotopic exchange with seawater during its erosion. However, sample number 14 presents a similar value to the FTB samples (Fig. 6a), so its formation could be related to hydrothermal activity.

Finally, the dense crusts observed in ROV images could be texturally associated with the *high-density silt* facies identified in core GC16 (Figs. 2d, e, f, 7, 8c, d, 9). These two features could be related to diagenetic processes that caused the formation of dense crusts and the slight cementation of the matrix of the *high-density silt* GC16 facies. The mentioned process took place during the marked hiatus calculated, which coincides with an abrupt change in colour and density in that core. Therefore, this facies could represent an initial stage in the formation of the dense crusts. These facts also demonstrate the presence of a strong bottom current influence that erodes the sediments, exposing the dense crusts to the seafloor.

Conclusions

The identification of ferruginous tubular bioforms (FTB) of goethite distributed over the seabed of the high marginal platform has been interpreted as exhumed bioform-like structures. Firstly, the FTB were formed in reducing micro-environments inside burrows during early diagenesis, triggering the pyrite precipitation of the FTB within holes of tube-dwelling macrofauna.

Besides, some hydrothermal activity has been deduced due to the high values of $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic ratios obtained from the FTB in comparison to the typical modern values

of seawater, together with high values of radiogenic Sr and U. Fluid venting activity, along with erosional processes, could produce the framboidal goethite of the FTB as a result of pseudomorphism after pyrite. Afterwards, the FTB were exhumed and remobilised by erosional activity. Therefore, these ferruginous structures can be used as indicators of a vigorous bottom-hugging current that acts and erodes sediments in deep-marine environments.

The identification of authigenic minerals such as apatite, calcite and dolomite over the seabed supports the hypothesis of the erosional activity acting by intense bottom-hugging currents that erode and exhume the FTB. This erosional activity has also been confirmed by the facies classification and the low sedimentation rates obtained from a sediment core of the high marginal platform. Hence, this work allowed obtaining a comprehensive view of the formation environment of these bioform-like structures and their subsequent erosion and distribution over the seafloor. In addition, these results provide knowledge about sedimentary processes in ancient environments, whose formation mechanisms are still unclear, helping to their interpretation.

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