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Total number of authors:

11

Published in:

Marine and Petroleum Geology

Link to article, DOI:

[10.1016/j.marpetgeo.2016.04.019](https://doi.org/10.1016/j.marpetgeo.2016.04.019)

Publication date:

2016

Document Version

Peer reviewed version

[Link back to DTU Orbit](#)

Citation (APA):

Faÿ-Gomord, O., Soete, J., Katika, K., Galaup, S., Caline, B., Descamps, F., Lasseur, E., Fabricius, I. L., Saïag, J., Swennen, R., & Vandycke, S. (2016). New insight into the microtexture of chalks from NMR analysis. *Marine and Petroleum Geology*, 75, 252-271. <https://doi.org/10.1016/j.marpetgeo.2016.04.019>

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Accepted Manuscript

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PII: S0264-8172(16)30114-3

DOI: [10.1016/j.marpetgeo.2016.04.019](https://doi.org/10.1016/j.marpetgeo.2016.04.019)

Reference: JMPG 2537

To appear in: *Marine and Petroleum Geology*

Received Date: 23 September 2015

Revised Date: 17 March 2016

Accepted Date: 20 April 2016

Please cite this article as: Faÿ-Gomord, O., Soete, J., Katika, K., Galaup, S., Caline, B., Descamps, F., Lasseur, E., Fabricius, I.L., Saiag, J., Swennen, R., Vandycke, S., New insight into the microtexture of chalks from NMR analysis, *Marine and Petroleum Geology* (2016), doi: 10.1016/j.marpetgeo.2016.04.019.

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NEW INSIGHT INTO THE MICROTTEXTURE OF CHALKS FROM NMR ANALYSIS

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ABSTRACT

An integrated petrographical and petrophysical study was carried out on a set of 35 outcrop chalk samples, covering a wide range of lithologies and textures. In this study various chalk rock-types have been characterized, in terms of microtextures and porous network, by integrating both geological, sediment-petrological and petrophysical data, including porosity, permeability, low-field NMR (Nuclear Magnetic Resonance), MICP and specific surface area (BET) measurements. The data allow an in depth understanding of the NMR signal of chinks, with a focus on tight chinks, including all low reservoir quality chinks independently of their sedimentological and/or diagenetic history. The study aims to develop an NMR-based approach to characterize a broad range of chalk samples. The provided laboratory low-field NMR chalk classification can be used as a guide to interpret NMR logging data.

Based on the petrographical and petrophysical analysis, 6 groups of samples were identified, each of them characterized by a unique NMR signature : (1) micritic chinks, (2) grainy chinks, (3) cemented chinks, (4) marl-seam chinks, (5) argillaceous chinks and (6) silicified chalk. NMR T_2 distributions were linked to pore body size and T_2 logarithmic (T_{2lm}) was calculated. It is apparent that tight chinks, whether their characteristics are sedimentological or diagenetic, yield smaller pore body sizes ($T_{2lm} < 20$ ms), as well as narrower pore throats (average radius < 150 nm) and lower permeability values (typically below 0.2 mD). Grainy chinks possess T_2 distributions reflecting larger pore sizes ($T_{2lm} > 60$ ms) and pore throats (average radius > 290 nm) and higher permeabilities (up to 13 mD). The marl-seam chalk samples yield bimodal T_2 distributions, with a first peak related to the micritic matrix pores and a second peak related to intraparticle pores within fossils. For all samples, permeability was inferred from NMR spectra using SDR (Schlumberger Doll Research) model.

Keywords: NMR, chalk, petrography, petrophysics, microporosity, porecasts, marl-seams, MICP

1. INTRODUCTION

Porous reservoir chalks have been thoroughly described and characterized in terms of petrography and petrophysics (Anderskov & Surlyk, 2011; Fabricius, 2003; Fabricius et al. 2007; Frykman, 2001; Hjuler & Fabricius, 2009; Jakobsen et al., 2000; Lindgreen et al. 2010). Mallon & Swarbrick, (2002, 2008) first focused on the large diversity of non-reservoir chalk lithologies that exhibit low permeabilities and suggested that these lithologies are present throughout much of the Chalk Group. According to the Joint Chalk Research group (Bailey et al., 1999), those non-reservoir or tight chalks include all chalks with matrix permeability lower than 0.2 mD, regardless of their origins. They became a focus of interest for petroleum industry since they might be underexplored reservoirs, or play a critical role in hydrocarbon migration, acting as seals or fluid conduits depending on their fracture patterns (Gennaro et al., 2013).

Recently, there has been a growing interest in the characterization of microtexture and pore network of microporous carbonate reservoirs (Brigaud et al., 2014; Cantrell and Hagerty, 1999; Deville de Periere et al., 2011; Kaczmarek et al., 2015; Lambert et al., 2006; Regnet et al., 2014; Richard et al., 2005; Vincent et al., 2011). Here, advanced techniques have been further developed and improved, including NMR, which gives detailed information about the porous network. The advantage of developing a NMR-based approach in chalk research is the application of NMR logging data, where NMR provides a continuous record of the pore size distribution in the reservoirs (Akbar et al., 1995, 2000; Fleury, 2000; Westphal et al., 2005). However, many effects affect the NMR signal such as pore coupling (Anand and Hirasaki, 2005) or surface relaxivity variation with mineralogy (Keating and Knight, 2012). These effects make the interpretation of NMR signals in carbonates a challenge, which was effectively addressed in a few studies (Brigaud et al., 2014; Vincent et al., 2011; Yousef et al., 2011).

NMR signals have been investigated in reservoir chalk pore networks; (Li et al., 2014; Megawati et al., 2012; Nurmi & Standen, 1997), but tight chalks have been disregarded to some extent. In order to enable the use of NMR logging for chalk rock-typing, this study aims to explain the NMR signal of a wide range of outcrop chalk samples, including tight chalks, based on petrographical observations and petrophysical measurements. Chalk samples were selected from North-West European outcrops and include chalks from various depositional settings, and exhibiting various diagenetic histories. As a result, a broad spectrum of mineralogies, degrees of cementation, compaction and sedimentary textures, and therefore a large spectrum of chalk petrophysical properties is covered in this study. Relationships are established between porosity, permeability, MICP, petrography and SEM observations, in order to determine the geological characteristics which constrain the NMR response.

2. MATERIAL AND METHODS

2.1. Sampling

In the field thirty-five core samples, 10 cm diameter by 30 cm long, were drilled from representative outcrops that reflect various sedimentological, tectonic and diagenetic histories, resulting in chalks displaying a wide range of lithologies, reservoir properties and petrographical textures. The samples were collected from 15 different locations spread on 6 study areas across Belgium, England and France (Table 1). The petrographical and petrophysical properties of each sample are the combined result of its sedimentological setting and a set of diagenetic modifications. This study focuses on the petrophysical and petrographic characteristics of the chalk lithologies rather than on the processes which led to the current reservoir properties.

Upper Cretaceous chalk samples ranging from the Cenomanian to Campanian were studied. Sedimentological features impacting the microtexture and petrophysical properties of chalks include insoluble residue content (e.g. Fabricius & Borre, 2007; Røgen & Fabricius, 2002; Strand et al., 2007), size and shape of chalk particles (e.g. Neugebauer, 1974, 1975) and the mode of deposition (e.g. Damholt & Surlyk, 2004; Scholle et al., 1998). In addition to these sedimentological features, diagenesis is controlled by local tectonics (e.g. Hancock, 1975; Jones et al., 1984; Mimran, 1975, 1978), the pore water chemistry (e.g. Fabricius & Borre, 2007) and burial setting. The latter, which includes the burial temperature, stress and pressure at a given depth, is a parameter with major impact on the diagenesis, that primarily governs the overall porosity reduction and thus strongly impacts the rock properties and textural characteristics (e.g. Borre & Fabricius, 1998; Fabricius, 2003; Scholle, 1974, 1977). Therefore outcrops with maximum burial paleodepths ranging from 200 m in Harmignies Quarry, Mons (Dupuis and Vandycke, 1989) to 1800 m in Flamborough Head, Yorkshire, UK (Menpes and Hillis, 1996) were selected (Figure 1).

2.2. Petrography

The homogeneity of the core samples was verified with X-ray CT scanning, using a Siemens somatom scanner with a resolution of 0.2 mm. Heterogeneous features like marl-seams, bioturbations or veins were not avoided while making plugs, since they are representative elements of the samples. In the laboratory, plugs (2.5 cm diameter by 5 cm long) were cored. Petrophysical and petrographical analyses were performed on the same plug whenever possible, or at least on nearby material.

Macroscopic samples were described using the classification established by Fritsen et al. (1996) who characterized 18 chalk lithotypes. Fritsen et al. (1996) modified Dunham's classification to make it suitable for application by the unaided eye. This means that the maximum mud particle size of 20 microns, as defined by Dunham (1962) or Lucia, (1995) could not be maintained. Several wackestone - observed as wackestones in thin-section - were classified as mudstones macroscopically.

For this study, 21 standard 30 μm -thick thin-sections were prepared. The sediment texture was described based on Dunham's (1962) classification of limestones, using a natural light microscope. JMicroVision Image analysis (Roduit, 2007) was performed on natural light thin-section images in order to quantify the proportion of grains ($> 20 \mu\text{m}$) present in the micritic matrix. Thin-sections were impregnated under vacuum conditions with a fluorescence epoxy resin to highlight the porous network. The pores recognizable by optical microscopy essentially correspond to intraparticle mesoporosity (Fabricius et al., 2008; Nurmi & Standen, 1997) since micropores are too small to be identified under the optical microscope. Image analysis, performed on fluorescence light thin-section images provides a quantification of this mesoporosity.

Scanning Electron Microscopy (SEM) on fresh surfaces of the samples was used to observe non-carbonate minerals, to identify microporosity and to study microtextural features. Investigations were carried out on an EM XL30 FEG field emission microscope equipped with an energy dispersive spectrometer (EDS) with image magnifications from 1000 to 20000 times. Classifications based on the morphology of the micritic matrix have been developed for microcrystalline calcite (Deville de Periere et al., 2011; Kaczmarek et al., 2015; Lambert et al., 2006). If nano-bioclots were sometimes observed among these microcrystalline calcites; they however never exceeded 5% of the sediment volume. The proposed classifications are therefore not applicable to chalk samples, and a detailed descriptive chalk microtextures classification needed to be developed. Throughout this paper, the term *microtexture* describes the texture of the chalk matrix, as observed under SEM; while *texture* always refers to the Dunham sedimentary texture classification.

In the frame of this study, a chalk diagenesis index was defined based on SEM observations. The diagenesis index ranks the samples on a scale from 0 to 10, with zero (0) being the uncemented chalk and ten (10) being the completely cemented chalk. Seven criteria are used to quantify the degree of diagenesis from SEM observation. They include parameters used for the classical micrite classification such as (1) the micritic matrix texture, grading from microrhombic to anhedral compact or (2) the grain contact from punctate to coalescent. Other parameters, more specific to chalks are (3) the disintegration of coccoliths, (4) coccolith grain overgrowths, (5) large cemented zones, (6) authigenic cement crystals and (7) intraparticle cementation, which is observed also in thin-section with fluorescence light. Each criterion was rated from 0 (uncemented) to 10 (completely cemented) and the average of those criteria provides the diagenesis index.

SEM was also used to scan and investigate porecast surfaces. Porecasts were obtained by dissolving 30 μm -thick thin-sections impregnated with epoxy-resin in a 10M hydrochloric acid solution, for 3 hours (Beckett and Sellwood, 1991). The study of porecasts is a helpful technique to visualize and evaluate the shape and distribution of the pore network.

Acid-insoluble residues of the samples were obtained by dissolving roughly crushed samples of chalk in 2M hydrochloric acid followed by washing of the residue with deionized water (Hjuler and Fabricius, 2009).

2.3. Petrophysics

2.3.1. Porosity and Permeability

The porosity was determined by direct measurement of grain volume and bulk volume. The grain volume was determined by helium expansion in a Boyle's Law EPS Porosimeter. The gas permeability of the samples was determined with a Vinci nitrogen permeameter. The plugs were mounted in a "Hassler" type core holder at a confining pressure of 400 psi and a steady state gas flow was established through the samples. The permeabilities were corrected for gas-slippage using Klinkenberg's empirical correlation.

2.3.2. Nuclear Magnetic Resonance (NMR)

The principles of the NMR method are described in detail by Grunewald & Knight (2009), Kenyon (1997) and Vincent et al. (2011). NMR is a powerful tool to characterize the pore size distribution, based on the magnetization decay of fluid molecules. NMR measurements consist of aligning the hydrogen nuclei (proton) with static and oscillating magnetic fields and then measuring the relaxation time after the removal of the magnetic field. The NMR relaxation response of a single fluid filled pore can be expressed by the exponential decay of the transverse magnetization signal, called $1/T_2$. T_2 is a characteristic time that represents the magnetic decay of the protons within the fluid after their polarization. The T_2 time of a single fluid filled pore is proportional to the volume to surface ratio (V/S) of the pore, and therefore, to the pore body size according to equation 1 (Coates et al., 1999).

$$1/T_2 = 1/T_{2(b)} + \rho (S/V) \quad (\text{Equation 1})$$

with T_2 : T_2 surface relaxation time, $T_{2(b)}$: bulk T_2 surface relaxation time, ρ : T_2 relaxing strength of the grain surface, S : surface area of the pore and V : volume of the pore.

The T_2 relaxation time distribution of all pores in a rock sample may be considered equivalent to the pore body-size distribution of the rock, although tortuosity and pore shape also strongly impact the T_2 distribution (Kenyon, 1997).

As part of this study, laboratory measurements were performed on 35 fully water-saturated 1 inch-plug samples, with a GeoSpec2 NMR Core Analyzer at 2.25MHz, in a temperature controlled chamber at 35°C and atmospheric pressure. T_2 relaxations were measured using the Carr-Purcel-Meiboom-Gill (CPMG) pulse sequences (Dunn et al., 2002). The polarization time was selected at 5 s, number of echoes at 16000, and the echo spacing (TE) at of 100 μ s. The T_2 relaxation spectra were generated using the WinDXP (Oxford Instruments, UK) software. All measurements were conducted at high

Signal to Noise Ratio (SNR>200) in order to increase the reliability of the results. For a detailed description of the analytical methods, the readers should refer to Katika et al., 2014.

2.3.3. Mercury-Injection Capillary Pressure (MICP)

Mercury-injection capillary pressure (MICP) tests allow the measurement of the distribution of the pore throat radii. The technique, first described by Purcell (1949) and subsequently by Wardlaw & Taylor (1976) is based on the principle that mercury is a near-perfect non-wetting liquid, that will only infiltrate the rock pores when force is applied. Spaces with larger pore throats are saturated at low pressure, but mercury progressively invades pores with smaller pore throats as pressure is step-by-step increased up to 30,000 psi. When a sufficiently high pressure is reached, the entire connected pore system, representing even the smallest pore throats, will be completely saturated. The pore throat radii distribution is calculated from the mercury capillary pressure curves by means of the Young Laplace's mechanical equilibrium equation. Measurements were performed on a selection of 21 samples, with a Micromeritics Autopore IV device whose accuracy ranges from 3.5 nm to 0.5 mm pore throat radii.

2.3.4. Specific surface (BET)

BET-analysis (Sing et al., 1985) on 8 carefully selected samples was carried out to get a better insight into the specific surface of peculiar samples. Analyses were performed with a Micromeritics ASAP 2020 instrument. BET-analysis provides specific surface area evaluation by nitrogen multilayer adsorption measured as a function of relative pressure. The technique encompasses external area and pore area evaluations to determine the total specific surface area in m^2/g yielding important information on the sample microtexture. In this study, the technique was used in order to estimate the specific surface area, and its impact on the NMR T_2 response was considered.

3. RESULTS

Based on the integration of both petrographical observations and NMR measurements, 6 chalk lithotypes were defined: (1) micritic chalk, (2) grainy chalk, (3) cemented chalk, (4) argillaceous chalk, (5) marl-seam chalk, and (6) silicified chalk (see overview of petrographical and petrophysical results in **Error! Reference source not found.** and **Error! Reference source not found.**).

3.1. Petrography

Cores have been sampled with carefully avoiding the obviously weathered zones, affected by cryogenic weathering or by meteoric water circulation (e.g. around fractures, damaged zones etc.). Duperret et al. (2005) showed that salt-weathering may affect the first centimeters of the seashore cliff outcrops. However, computerized tomography of the 30-cm long core samples showed no variations in density within each sample, reflecting no differential weathering. In addition, SEM observation of the 35 samples showed no presence of precipitation related to meteoric diagenesis as described by Hjuler and Fabricius (2009), and very rare halite crystals precipitations within sea-shore samples. Hence, in this study, the impact of surface diagenesis is considered as minor.

3.1.1. Micritic chalk

Micritic chalks were named based on their mud-dominated texture, often mudstone, occasionally wackestone, according to Dunham's (1962) classification (**Error! Reference source not found.** and Table 2). Macroscopically, micritic chalk is a pure (less than 1% insoluble residue) chalk mudstone, which is not strongly indurated. CT scanning showed that micritic chalks are also very homogeneous samples. Thin-section observations showed mostly the presence of mudstone fabrics, where grains are mostly planktonic foraminifera which constitute only 1 to 6% of the samples based on image analysis. These mudstone facies form in deep offshore depositional environments, with low hydrodynamics and particles settling in the water column (Lasseur et al., 2009). SEM observations clearly show that the main components of the rock are very well preserved coccolith fragments. The fragments gave rise to a microrhombic matrix texture, with rare authigenic calcite crystals. Contacts between coccolith fragments are mostly punctate and the interparticle porosity is well preserved. The diagenesis index of micritic chalk was determined under SEM and fluorescence light microscopy ranged from 0 to 2, indicative of uncemented to poorly cemented samples. SEM observation of resin porecasts show that pores form a well-connected network of spherical pores. High magnification SEM-photographs of the outer edges of a foraminifera test show the very fine connections between the intraparticle porosity and the surrounding matrix pores.

3.1.2. Grainy chalk

Grainy chalk samples are grain-dominated and possess a packstone texture. Macroscopically, grainy chalks cannot always be easily distinguished from micritic chalks, as they also appear similar to low strength pure chalks. They often appear white, homogeneous and according to the classification of Fritsen et al. (1996) most of them qualify as mudstone in terms of macroscopic texture. Only one coarse lag facies sample infilling the base contourite channelized structures on the Normandy cliffs (Lasseur, 2007) can be classified as grainy chalk with the naked eye. The other grainy chalk samples were characterized as such based on microscopic observations. In thin-section, those chalks show a higher foraminiferal and bioclastic content which, as described by Nurmi & Standen (1997), gives rise to a packstone texture (Dunham, 1962), with 32 to 43% of grains occurring in a micritic matrix. Under SEM, the micritic matrix of grainy chalks is rather similar to the micritic chalk matrix. The diagenesis is equally low ranging from 1 to 2.

3.1.3. Cemented chalk

The cemented chalk lithotypes include chalk samples cemented during either eodiagenesis or mesodiagenesis. All cemented chalk samples are macroscopically characterized by their high strength, which either comes from early cementation in hardgrounds, burial diagenesis or a combination of both. In thin-section, cemented chalks display a mudstone texture, with 5 to 8% grains, which are (mostly forams or calcispheres), which are either cemented with large sparite crystals of several dozen micrometers or micritized. Although they rank as Dunham's mudstones, the sedimentological origin of cemented chalk deposits varies widely. SEM suggests that cementation obliterates sedimentological differences among the cemented chalk samples. All cemented chalks exhibit similar microtextures with coalescent grain contacts, many authigenic calcite crystals and grain overgrowths. Microtextural differences were rarely encountered between early cemented chalks and stylolitized chalks, cemented through deep burial diagenesis (e.g. Flamborough Head samples). Deeply buried chalks show a higher disintegration of coccoliths and seemingly more grain overgrowths than early cemented chalks. Cemented chalk resin porecasts indicate a porous network with straight-edged pores under SEM, generated by the euhedral crystal faces of calcite rhombodra. Pore throats also appear very narrow and void zones of $>10\ \mu\text{m}$ in the porecast reflect large grain aggregates, cemented together, completely filling the pores. Overall, the diagenesis index values of cemented chalks range from 6 to 7.

3.1.4. Marl-seam chalk

Marl-seam structures have been observed and described in chalk since Weiler (1957), and are commonly described as common pressure solution features, resulting from burial diagenesis (Lind, 1993), although a strict sedimentary origin is still considered by some authors (Wray & Jeans, 2014). According to Jennings & Mazzullo (1986), the minimal burial depth required to form marl-seams in chalk is 400m. Marl-seam samples are often associated with a pseudo-nodular fabric and the thickness

and geometry of the marl-seams depend on the initial chalk lithology as well as the intensity of the pressure solution processes.

Although marl-seams may preferentially initiate in a layer with a slightly higher initial clay-content, field observations show that marl-seams form essentially in homogeneous chalks with low insoluble residue (< 5%). Marl-seams are not observed in chalks with high insoluble residue (> 10%), nor in chalks with regular chert nodule bands nor in hardgrounds. Computerized tomography analysis revealed that marl-seams density is higher than surrounding chalk (Figure 5). Microscopic observations reveal that marl-seam chalks possess a mudstone – wackestone texture, with a percentage of grains ranging from 6 to 17%. Fluorescence microscopy highlights the presence of intraparticle pores, inside calcispheres, foraminifera and other bioclasts.

SEM observations of marl-seam chalks show a wide range of microtextures, which relate to the intensity of the burial pressure solution or the initial sedimentary clay-content. However, sampling is also critical because of the heterogeneity of the samples. For example the microtexture observed inside marl-seams is significantly different from the surrounding chalk. The sampling of marl-seams for insoluble residue measurements also strongly influences the measurements, which range from 2 to 15%. Unlike other chalk lithotypes, there is not one typical texture that allows to identify marl-seam chalks. If the sampling is made in the clay-rich seams, then the microtexture is dominated by the clay flakes, which may show a preferential orientation if the sampling is made a few millimeters away from the clay-seams, the texture is not dominated by the clay-content and microtextural features includes authigenic cement crystals and grain overgrowths that do not develop in the clay-rich seams.

3.1.5. Argillaceous Chalk

Argillaceous chalk samples come from Cap-Blanc Nez, (Boulonnais, France) and Eastbourne (Sussex, England) and are of early Cenomanian age. The argillaceous facies is clearly identifiable macroscopically by its light to dark grey color. Burrowings are very common, often from *Zoophycos* and *Chondrites* (Amédéo & Robaszynski, 2001). However, computerized tomography revealed an homogeneous density within the samples (Figure 5). The insoluble residue content of argillaceous chalks ranges from 10% to 30% and is mostly detrital, including clays (illite, smectite, glauconite or kaolinite), quartz and a small amount of pyrite. One sample from Cap Blanc Nez (OF/CB10, Table 2) shows lower insoluble residue because it had been affected by early diagenesis, characterized by an extensive burrowing. Early calcite cementation likely reduced the relative abundance of insoluble residue. The Cenomanian coincides with the beginning of a major transgression leading to the formation of the Chalk sea. The detrital input was still high then, whereas the biological carbonate production from coccolithophorids was probably moderate. The shift from dominance of detrital input to carbonate production lead to the sedimentation of argillaceous chalks, with petrographic characteristics somewhere in between marls and chalk.

Thin-section observations showed that clays are dispersed in the brownish matrix. The texture of argillaceous chalk varies from mudstone to wackestone, where grains mostly include foraminifera, bioclasts and calcisphere, representing between 7 and 18% from thin-section image analysis. However, argillaceous chalk are clearly recognizable by their microtexture under the SEM. Clay flakes are dispersed in the matrix, and under the mechanical compaction stress they tend to align parallel to the bedding, as described by Mallon & Swarbrick (2008). The texture of the matrix appears more compact in argillaceous chalk than in micritic chalk, because of the poor sorting. In contrast, pure micritic chalks likely develop a more solid framework by crystal overgrowth cementation during diagenesis, so they better withstand mechanical compaction. Because of the dispersed clay flakes in the matrix, contacts between coccolith fragments are reduced and argillaceous chalks develop very low grain-to-grain contact dissolution or grain overgrowths. Thus, even if they are initially more affected by mechanical compaction, chemical compaction is lower than in clay-free chalks. In the determination of the diagenesis index, criteria such as crystal overgrowths or coalescence of grain contacts are very low, while the overall micritic texture and the coccolith disintegration are significantly higher than in micritic chalks. The diagenesis index values range from 3.5 to 5 for argillaceous chalk and slightly higher, with values up to 6, for chalks having undergone early cementation, such as firmgrounds.

3.1.6. Silicified Chalk

Silicified chalks are not often encountered in chalk outcrops, where silica is mostly present under the form of chert nodules. Silicified chalks have been previously described in the Ekofisk and South Arne oil fields in the North Sea (Jakobsen et al., 2000; Lindgreen et al., 2010; Gennaro et al., 2013). The only studied silicified outcrop chalk sample in this study comes from Brunneval, in Normandy (France). In the field, this silicified chalk appears as a 30 cm thick bed and displays a clear silicification front. The light-grey silicified rock still possesses a chalk texture with distinct bioclasts, but the induration of the rock is higher than the surrounding chalk. The insoluble residue quantity of this sample reaches 76%.

Fluorescence microscopy revealed zones of varying porosity. In each of these zones, the initial chalk texture was preserved with bioclast and calcisphere outlines still distinguishable. In the higher porosity zone, patches of amorphous silica occur; while in the lower porosity zone, which appears completely silicified, chalcedony can be observed in what used to be the chalk-matrix.

SEM micrographs from thin-sections show that silica is dispersed in the chalk matrix but is developed as a cement in foraminifera and calcisphere chambers (**Error! Reference source not found.**). Furthermore, SEM observations of rock chips indicate that the silica is present as both chalcedony aggregates and as subhedral to euhedral quartz crystals (Figure 6). The precipitation of nano-quartz has been interpreted as a consequence of the supersaturation of SiO₂ in pore water (Jakobsen et al., 2000; Fröhlich, 2007; Lindgreen et al., 2010), related to increased silica concentrations, most probably

resulting from the dissolution of biogenic silica (i.e. radiolaria, diatoms and silicoflagellates). However, this proposed direct precipitation of quartz is highly controversial because studies based on X-ray diffraction and SEM-based petrography indicate that biogenic silica dissolves and re-precipitates as opal-CT, which after sufficient time and temperature transforms into quartz (Kastner 1981). The nano-size of the resulting quartz aggregates mimics that of Opal-CT lepispheres (Hjuler and Fabricius 2009).

3.2. Petrophysics

3.2.1. Porosity and Permeability

Petrophysically, chalk is defined as a mineralogically pure, highly porous and low permeability carbonate rock. Among selected outcrop samples, micritic chalk ranges from 30 to 45% porosity and 2 to 6 mD permeability and grainy chalk shows similar values with porosity ranging from 30 to 45%, and permeability from 3 to 13 mD. The atypical, highly-permeable sample of 13 mD permeability (OF/ETR33, Table 3) is a grainstone-textured bioclastic chalk from Etretat (France).

However, not all chalks are porous, nor can they all be considered pure carbonate. Some chalks show exceptionally low permeability values (Mallon and Swarbrick, 2008, 2002; Gennaro et al., 2013), which will strongly affect the overall reservoir properties. The Joint Chalk Research (JCR, <http://jointchalkresearch.org/>) has termed those low reservoir chalks as “tight chalks”, which are characterized by a matrix permeability below 0.2 mD. In the studied samples, all the argillaceous chalks, the deeply-buried cemented chalk from Flamborough Head (England, samples OF/FA15, OF/FA39 and OF/FA11), the nodular marl-seam chalk from Cap Blanc Nez (OF/CB16) and the silicified chalk (OF/BR01) samples qualify as tight chalks, because of their very low permeability values (**Error! Reference source not found.**). As mentioned before, marl-seam exhibit variations in the intensity of compaction. That results in porosity variations from 14 to 40% and permeability from 0.19 to 2.7 mD. The cemented chalks from Cap Blanc Nez (OF/CB14) and Normandy (OF/SC01) show higher porosity and permeability, which may be explained by the lower maximum burial paleodepth, not exceeding 500 m. The determination coefficient (R^2) between porosity and permeability values for all studied samples equals 0.78 (**Error! Reference source not found.A**).

3.2.2. MICP

Many case studies used MICP to define micropores, mesopores and macropores in carbonates based on the pore throat radius (PTR) (Bousquié, 1979; Frank et al., 2005; Goni et al., 1968; Skalinski & Kenter, 2015). Each study proposed its own cut-off values to define micropores, ranging from a PTR lower than 30nm (Goni et al., 1968) to lower than 250 nm (Frank et al., 2005). For the studied chalk samples, PTRs range from 12 to 550 nm (**Error! Reference source not found.**). Measurements show an average PTR of 255 nm for micritic chalks and 380 nm for grainy chalks. The actual PTRs of pores

in micritic chalks and grainy chalks overlap since sedimentary texture and NMR measurements were used to differentiate between chalk lithotypes. Due to overgrowths and formation of euhedral calcite crystals, PTRs in cemented chalk are reduced to an average of 120 nm. The reduced pore throat size of cemented chalk can be clearly understood and visualized with the epoxy resin pore-casts (**Error! Reference source not found.**). Argillaceous chalks are characterized by an average PTR of 50 nm, with pore throats size reduced by the presence of thin clay flakes in the pore throats. The smallest average PTR (12 nm) is observed in the silicified chalk sample. In the marl-seam chalks PTRs measurements range from 50 to 215 nm, depending on the compaction intensity and on the microtexture of the chalk before the initiation of burial diagenesis.

The pore throat radii distribution is unimodal and normally distributed for micritic, cemented and silicified chalk, suggesting that one family of pore throats controls mercury intrusion into the studied samples. Pore throat radii distributions of argillaceous chalks are unimodal with a significant shoulder towards low values, also observed in some marl-seam samples. Pore throat distributions of grainy chalk samples OF/CM06 and OF/SS01 are unimodal and symmetrical, but OF/ETR33 (**Error! Reference source not found.**) displays a bimodal pore throat distribution with one dominant peak (0.1-5 μm) with a shoulder towards high values and a small broad peak at high values (10-100 μm), which may result from a second family of pore throats, related to the larger amount of coarser grains in the samples, accompanied by inter- and intra-particle porosity.

3.2.3. BET measurements

The specific surface of 8 representative samples (2 micritic chalks, 1 grainy chalk, 2 cemented chalks, 2 marl-seam chalks, 1 argillaceous chalk) was measured. The specific surface of micritic chalk samples was measured at 2.2 m^2/g , which similar to the data reported by Røgen & Fabricius (2002), stating that the specific surface area of chalk biogenic calcite is on average 2 m^2/g . The latter authors also state that the specific surface area is mostly controlled by carbonate content and mineralogy of the non-carbonate fraction and that the specific surface area of quartz is about 5 m^2/g , that of kaolinite about 15 m^2/g and that of smectite about 60 m^2/g . The high specific surface areas for the non-carbonate mineralogies explain the higher values measured for argillaceous chalk (OF/CB11) and marl-seam chalk (OF/CB13 & OF/CB16), respectively 11.23 m^2/g and 8.6 m^2/g (**Error! Reference source not found.**). For the examined chalk samples, this is consistent with the clay flakes dispersed in the matrix that were observed with SEM. The specific surface of cemented chalk equals 1.9 m^2/g for the Flamborough Head deeply-buried chalk sample (OF/FA39) and 2.7 m^2/g for the Cap Blanc Nez (France) firmground sample (OF/CB14). Based on the insoluble residue amounts of respectively 3.5% and 5.9% of the latter samples, higher specific surfaces were expected. Overgrowth cement precipitation on the surface of the grains however smoothed the particles and reduced the specific surface area (Borre & Fabricius, 1998; Olsen et al., 2008), which is well illustrated by the porecasts

(**Error! Reference source not found.**, B). The impact of cementation and insoluble residue counteract each other, resulting in specific surface area values similar to micritic chalks. The lowest specific surface area ($0.8 \text{ m}^2/\text{g}$) was measured on an atypical grainstone-textured chalk sample (OF/ETR33) and is coherent with presence of coarser grains.

3.2.4. Low-field NMR

The T_2 distribution reflects a distribution of V/S ratios, which includes the pore shape factor as well as the surface rugosity. The high specific surface provided by clays in argillaceous or marl-seam chalk therefore tends to decrease the average T_2 time. The logarithmic mean of the T_2 distribution (T_{2lm}) was calculated for each sample from the unimodal T_2 time distribution. The average T_{2lm} value equals 47 ms for micritic chalks and 81 ms for grainy chalks. As expected, the average T_{2lm} of argillaceous chalk (5 ms) is significantly lower, because the dispersed clay flakes in the micritic matrix both effectively reduce the pore volume and increase the specific surface area so that T_2 time decreases. The low value of T_{2lm} in cemented chalks (16 ms) relates to the pore size reduction due to cementation.

Sediment deposition in strong hydrodynamic environments results in coarser chalk, characterized by a packstone texture and larger pores. Both factors lead to higher T_{2lm} values in NMR. The OF/ETR33 sample displays a bimodal MICP distribution, which might reflect the presence of a second family of pore throats, related to larger grains. This second family of pores is confirmed by a bimodal T_2 distribution in NMR with a second peak towards higher T_2 values (800-3000 ms) and is in agreement with thin-section observations showing large inter- and intraparticle pores related to large sponges or bryozoa fragments.

Marl-seam chalks often show unimodal MICP patterns (**Error! Reference source not found.**A) while in NMR clear bimodal T_2 distributions are observed (**Error! Reference source not found.**B). The clay-rich seams and the clean chalk layers in marl-seam chalks are both part of the highest peak of the NMR- T_2 distribution, which is referred to as matrix porosity. The concentration of clay flakes in the matrix, along preferential seams, locally reduces pore size and thus also T_2 values but they do not isolate the pores from the surrounding cemented and clay-free matrix, resulting in a homogeneous first peak signal in NMR, referred to as matrix peak. Based on petrographical observations, the high- T_2 value lowest peak of the NMR signal is related to the high intraparticle porosity featured in marl-seams layers. The unimodal MICP signal can be explained by the fact that the scattered and isolated intraparticle pores are only accessible through narrow pore throats in the foraminifera tests (**Error! Reference source not found.**C). On the contrary, due to the large difference in pore size between matrix porosity and intraparticle porosity and because of a weak coupling-effect, the NMR signature is slightly bimodal. The T_{2lm} of bimodal marl-seam chalk samples have been calculated for the first mode of the T_2 distribution only and therefore reflects the log mean of the matrix porous network, excluding

the intraparticle pores. A relationship between the insoluble residue content and the T_{2lm} value is visible for the marl-seam samples (**Error! Reference source not found.**D), and shows that pressure-solution results in higher clay concentration and lower T_{2lm} values.

4. DISCUSSION

Following a detailed and integrated analysis of available petrophysical and petrographical data, it appears that NMR T_2 distribution provides invaluable information about chalk porous network. Five lithotypes can be directly inferred from low-field NMR T_2 distributions and the permeability of each sample can be assessed. The NMR signature records key-information about chalk microtexture that results from both sedimentological and diagenetic history.

4.1. Marl-seam samples

Marl-seam samples stand out from other chalk lithologies by their wide range of petrographic and petrophysical properties. The microtexture of marl-seam chalk is by far the most complex because of the initial intrinsic heterogeneities and pressure dissolution intensity that affect the characteristics of each sample. This lithotype is formed by alternating poorly cemented clay-rich seams and clean chalk intervals whose pores are more cemented and therefore a wide range of microtextures and pore features are observed. Despite these heterogeneities, all marl-seam chalk samples have one common characteristic: a bimodal NMR T_2 distribution (**Error! Reference source not found.**).

The T_2 values of the first peak have a range that falls between typical values for argillaceous and clean chalks (5-100 ms), while the second peak is on the coarser side of chalk T_2 distributions (> 100 ms). It was tempting to attribute the first NMR-peak to clay rich-seams and the second to clean chalk. However, the integration of NMR measurements with petrographical observations led to the conclusion that the second mode of porosity is in fact related to the well-preserved intraparticle porosity in those samples (see 3.2.4, **Error! Reference source not found.**). All the matrix pores were therefore registered in one single broad peak, which might be inferred to their very good connectivity and possibly a pore coupling effect. The preservation of the intraparticle porosity in marl-seam layers is interpreted as resulting from a diagenetic shielding effect related to the presence of the clays. The shielding effect induced by clay flakes also reduces the grain to grain contacts and subsequently decrease dissolution processes, inhibiting calcite cementation.

Westphal (2006) described the differential diagenesis in marl-seams with calcium-carbonate export from the marl-seam layers and import in the pure layers. In this study, marl-seams layers (OF/CB13, OF/SC02, OF/SC03) are often found in between cemented chalk layers (OF/CB14, OF/SC01) with especially low percentages or completely lacking intraparticle porosity, while cementation is low to nonexistent in marl-seams layers, potentially because of a shielding effect. Note that this shielding

effect may have been of influence already before the formation of marl-seams caused by burial diagenesis. While marl-seams initiation may be sedimentologically-controlled, and generated on bedding-parallel inhomogeneities; their atypical bimodal NMR signal is likely the result of a differential diagenesis. The unique bimodal signal of marl-seams is likely related to interparticle porosity preservation, because marl-seams layers provide calcium carbonate to the system by pressure-solution processes without in-situ cementation; and fluids saturated in calcium-carbonate will preferentially circulate and precipitate in pure chalk layers.

4.2. The peculiar silicified chalk

NMR and MICP signals of each NMR-unimodal chalk lithotype are compared (**Error! Reference source not found.**) and the micritic, grainy, cemented and argillaceous chalks have unique T_2 distributions. Only the silicified chalk sample (OF/BR01) overlaps with the cemented chalks. Pore throat sizes of the silicified chalk sample are, however, lower than any other lithotype and yield very low permeability (0.04 mD). Pore throat size and permeability are seemingly more reduced by silicification than pore volume and overall porosity (26.4%). The nanometer-sized silica particles, which are dispersed in the matrix, probably obstruct pore-throat accesses, while neither drastically reducing pore volumes. Silicified chalk beds occur as centimeter-thick layers, and may act as intra-reservoir seals (Gennaro et al, 2013; Jakobsen et al., 2000; Lindgreen et al., 2010; 2012). NMR-logging in oil-field will not be sufficient to discriminate between cemented chalk and the silicified layers, but an integrated study with gamma-ray logging and/or SEM cutting observations will normally allow to identify them.

4.3. Rock typing from NMR

The overview of this integrated petrographical and petrophysical catalog of chalks shows how sedimentary setting and burial history impact the chalk properties, affect their microtextures and thus play a crucial role in shaping the NMR signal. The main parameters specifically controlling chalk microtextures and hence T_2 distributions are (1) the primary grain size, (2) the non-carbonate content, and (3) the degree of cementation.

Non-carbonate constituents and cements are the two key factors altering chalk reservoir properties, by reducing porosity and permeability. Thin clay flakes surrounding the grains as well as pore-filling authigenic crystals or grain-overgrowth cementation reduce both the pore throat and pore-body sizes (Figure 12). However, they may sometimes counteract each other, as for example when high detritic clay content prevents cementation during burial diagenesis.

Vincent et al. (2011) proposed a cut-off value between micro- and mesoporosity at a T_2 time of 200 ms in neritic marine carbonates. This threshold is not sufficient to distinguish the different pore types that

were observed in chalk samples. Based on the cut-off of the above mentioned authors, all samples in this study, except the grainy chalk, would be interpreted as 100% microporous. Therefore, new cut-off values were defined to separate three classes of pores in the studied chalk samples. The cut-off for chalk-microporosity was defined based on the micritic chalk samples, whose unimodal peak always plots between $T_2 = 25$ ms and $T_2 = 100$ ms. This T_2 interval defines the chalk microporosity. All values below $T_2 = 25$ ms were defined as chalk-nanoporosity, and values above 100 ms as chalk-mesoporosity.

The observed lithotypes plot in clearly separate areas based on the relative abundance of these porosity classes (Figure 13). In this diagram, the various chalk lithotypes that were previously described clearly stand out. The porous network of grainy chinks is made up of chalk-microporosity in the mud-matrix fraction and mesoporosity in the grain-fraction of the sediment. The micritic matrix porosity of marl-seam chinks ranges between the chalk-nanoporous and microporous poles, depending on the clay-content and degree of cementation in each sample. The second mode of the bimodal T_2 distribution materializes in the diagram by a shift of the pore class percentages towards the chalk-mesoporosity pole.

Chalk-nanoporosity is mostly represented in the cemented and clay-rich chinks, where the pore size is reduced. Nevertheless, the extremely low T_2 values of argillaceous chinks also reflects the increased specific surface due to clays, and the small amount of pyrite that was observed in argillaceous chalk also has a reducing effect on the T_2 time. However, the good correspondence between MICP and NMR responses (**Error! Reference source not found.**) validates the coherence of the acquired NMR responses.

To go further, uniaxial compressive strength has been measured on the studied samples and clear distinct behavior appeared for each NMR-determined chalk lithotype (Descamps et al., 2015), opening the prospect of predicting some geomechanical properties from NMR measurements.

4.4. Permeability estimation from NMR

Rock permeability can be estimated if the pore throat size distribution is known. Because the combined results of MICP and NMR in this study showed that pore throat and NMR pore body size are related, with a determination coefficient of $R^2 = 0.88$ (**Error! Reference source not found.**B, C), permeability can thus be inferred from NMR measurements in chinks (Minh et al., 1997).

The applicability of the Schlumberger-Doll-Research (SDR) model (Kenyon et al., 1988), used to calculate permeability from NMR measurements, was tested on studied chalk samples. In the K_{SDR} model, the permeability is predicted from the sample porosity (ϕ) and T_{2lm} (Eq. 2). The calibration constant 'a' was determined graphically from the gradient of the best fit curve to the data in the

$T_2(1m)^2 \phi^4$ versus permeability plot (**Error! Reference source not found.A**) and equals 0.09. SDR relationships in carbonates are expected to yield low values for 'a' (Minh et al., 1997). Furthermore, smaller pores generally contain much more surface area and higher surface area to volume ratios, which results in shorter surface relaxation times according to Equation 1 (Coates et al., 1999). The absence of macropores in the chalk samples results in a low value for the calibration constant 'a'.

$$K_{SDR} = a (T_{2lm})^2 \phi^4 \quad (\text{Equation 2})$$

with ϕ : porosity, a: calibration constant, $T_2(s)$: T_2 surface relaxation time, ρ : T_2 relaxing strength of the grain surface, S: surface area of the pore and V: volume of the pore.

The pore-coupling phenomenon which may occur in grainy chalk directly impacts the T_{2lm} average of the T_2 distribution and is a key parameter for calculating the permeability from NMR data in the SDR model (Equation 1). Despite this potential inaccuracy, the SDR modelled permeability is in rather good agreement with measured core permeabilities. The relation between both permeability is described in equation 3 and yields a determination coefficient of 0.85 (**Error! Reference source not found.B**).

$$K_{core} = 1.2266 \cdot K_{SDR}^{0.5034} \quad (\text{Equation 3})$$

Knowing that the NMR based SDR permeability model is applicable to chalk, permeability can potentially be estimated from NMR well logging based on this calibration (Equation 3). However, it is important to remember that T_2 is sensitive to temperature. Indeed, physical processes such as the relaxation of the bulk fluid molecules, the molecules interaction with the surface, or the molecules diffusion between pores, could be affected by borehole temperature (Freedman, 2006). The latter processes depend on the rock properties and can result in an increase, decrease or null T_2 shift with temperature. NMR data should be corrected for temperature affects to reduce uncertainty in NMR-estimated permeabilities used in reservoir models.

5. CONCLUSION

On the basis of 35 chalk outcrop samples, covering a wide range of sedimentary facies, burial histories and petrophysical properties, this study provides keys for the interpretation of low-field NMR analyses, with regards to petrographical and petrophysical characteristics of chalks.

The main conclusions are :

- (1) Low-field NMR T_2 distributions allow 5 chalk lithotypes to be differentiated on the basis of their pore sizes and shapes: micritic, grainy, cemented, argillaceous and marl-seam chalks. Chalk NMR signals are mainly constrained by three parameters: the depositional grain size, the non-carbonate content and the degree of cementation. In more detail, we observed that:
 - in micritic chalks, clean and highly porous mudstones made essentially of poorly cemented coccolith fragments, T_{2lm} values range from 40 to 60 ms.
 - in argillaceous chalks, the high clay content results in a higher specific surface area and thus lower T_2 values, with T_{2lm} ranging from 3 to 10 ms.
 - in cemented chalks, cementation reduces pore size volumes and thus T_2 values, with T_{2lm} ranging from 15 to 18 ms.
 - in grainy chalks, characterized by a packstone texture and low cementation, NMR- T_2 distribution reflects the coarse porosity related to the high bioclastic fraction (foraminifera, echinoderms, bryozoan, etc.)
 - in marl-seam chalks, the NMR signature is characterized by a bimodal T_2 distribution. The main peak probably represents the pores of the matrix, whether or not they contain clays, while the second peak, with higher T_2 values most likely reflect the intraparticle porosity.
- (2) The preservation of intraparticle porosity in marl-seam chalks likely stem from a shielding effect caused by clays. Marl-seam chalks therefore may act as fluid barriers and the intermixed clay-free chalk behave as preferential fluid pathways. These diagenetic features therefore should be taken into account to understand the pore-collapse mechanisms occurring in some North Sea oil fields.
- (3) Tight chalks or low reservoir quality chalks encompass different lithotypes. The main permeability-reducing factors are: (1) the non-carbonate content, related to sedimentological settings (e.g. argillaceous chalks) and burial diagenesis (e.g. marl-seam chalks); (2) the degree of cementation, either eogenetic (e.g. hardgrounds) and relating to the depositional setting or mesogenetic and resulting from chemical compaction. Those parameters strongly modify chalk microtexture and reduce pore-throat and pore body sizes, altering permeability.
- (4) Permeability can be inferred from low-field NMR measurements using the SDR (Schlumberger Doll Research) model.

- (5) By applying the proposed NMR-based chalk classification, a continuous record of the pore size and chalk microtexture characteristics in the hydrocarbon reservoirs can be obtained.

NMR, together with MICP and SEM observations, provides detailed and holistic information about chalk pore networks, and a good insight into the chalk microtexture. NMR well logging, especially if combined with observations on cuttings should allow an accurate prediction of the petrological and petrophysical characteristics of chalks in wells. NMR is again proving to be a powerful tool for the analysis of pore networks, allowing the discrimination of lithotypes even in chalks.

6. ACKNOWLEDGEMENTS

This study has been funded by TOTAL SA, as part of a “Fractured Tight Chalk” project, including the PhD of Mrs Ophélie Faÿ-Gomord. Our thanks goes to Yves Leroy, TOTAL, project leader for the support he granted us throughout this study. We are thankful to DREAL Nord Pas de Calais and the Fortress of Mimoyecques for their sampling authorizations in protected areas, as well as CBR cement for the opportunity to work and sample in the Harmignies quarry (Belgium). We would also like to thank Herman Nijs (KU Leuven), for the preparation of the thin-sections and his smart input in the making of porecasts, and Evy Vereecken (KU Leuven) for her help and guidance during the MICP measurements. We also thank Pierre-Yves Collin (University of Burgundy) and Jean-Pierre Sizun (University of Besançon) for the sampling of certain outcrops in Haute-Normandy (France). We are very grateful to Pierre Van Landschoot (UMONS) for his help during different steps of sample preparation, as to Francis Amédéo (University of Burgundy) for his help and useful advice during fieldwork in Cap Blanc Nez (France). Finally, many thanks to Franciszek J. Hasiuk and Axel Munnecke for their constructive reviews which led to significant improvement of the manuscript.

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Figure 1: Map of the present day depth position of chalk deposits in North-West Europe and the location of the sampled outcrops with their maximum paleodepths (modified from Ziegler, 1990 and Hjuler & Fabricius, 2009).

Figure 1: Overview of petrographical and petrophysical results for micritic chalk, grainy chalk and cemented chalk. The T_{2lm} of the NMR T_2 distributions ranges from 37-58 ms for micritic chalk, 62-90 ms for grainy chalk, and 15-18ms for cemented chalk samples. MICP pore throat radius size distributions show average pore throat size ranging from 194-310 nm for micritic chalk, 296-540 nm for grainy chalk and 113-137 nm for cemented chalk.

Figure 2: Overview of petrographical and petrophysical results for marl-seam chalk, argillaceous chalk and silicified chalk. The T_{2lm} of the NMR T_2 distributions ranges from 8-32 ms for marl-seam chalk, 3-11 ms for argillaceous chalk, and 17.6 ms for the silicified chalk sample. MICP pore throat size distributions show average pore throat size ranging from 50-213 nm for marl-seam chalk, 38-61 nm for coarse chalk and 12 nm for silicified chalk sample. Note that the SEM microtexture of marl-seam chalk varies widely – two extreme cases are pictured here with on the left, the microtexture of a clean uncemented chalk interval, and on the right the microtexture of a clay-rich seam.

Figure 3: Dunham textures encountered for each chalk lithotype: thin-section observation of 3 representative samples from mudstone (OF/CH01), wackestone (OF/CB06) and packestone (OF/CM02) textures.

Figure 5: 3D Computerized tomography reconstitution of OF/CB06, an argillaceous chalk sample and OF/CB13, a marl-seams chalk sample. The homogeneity of sample OF/CB06 was assessed, only few bioclasts appear in white. In sample OF/CB13, the lower density of the marl-seams is reflected by a lighter grey seams (2) in a darker matrix (1).

Figure 6: Silicified chalk (sample OF/BR01) under SEM. (A) Overview of the samples with patches of amorphous silica. (B) Zoom on an amorphous silica zone (C) Micritic chalky matrix with some intact coccoliths, nano-aggregates of silica (D) Small quartz crystals between the grains. (E and F) Back-Scattered Electron SEM micrograph of a thin-section, with the darker zones being more silicified, frequently occurring as cement inside forams and calcispheres. Calcite of the shells is well preserved (white).

Figure 7: Petrophysical measurements with (A) porosity versus permeability, (B) Average PTR (Pore Throat Radius) versus permeability, (C) T_{2lm} versus average PTR, and (D) T_{2lm} versus insoluble residue displaying an exponential relationship.

Figure 8: Porecast (upper images) and rock chip (lower images) SEM pictures of micritic chalk (sample OF/CH01: A, C) and cemented chalk (sample FA39B: B, C). Porecasts from epoxy resin

allow to visualize the porous network, while rock sample pictures show the rock microtexture. The geometrical pore shapes of cemented chalk is highlighted by an arrow (B).

Figure 9: Grainy chalk OF/ETR33 coarse sample (A) MICP slightly bimodal pore-throat radii size distribution (B) NMR bimodal T_2 distribution. Blue curves are micritic chalk distribution, used as references. (C) Thin-section observation under transmitted light with a binocular. Red arrows show the porosity related to large bioclasts. (D) Back-Scattered Electron SEM-micrograph of a thin-section showing the matrix microporosity (red arrows).

Figure 10: Marl-seam sample (sample OF/CB13): (A) Unimodal MICP signal, (B) Bimodal NMR signal with the first peak related to the matrix and the second to the intraparticle porosity (C) Epoxy resin porecast showing the narrow pores in the forams connecting the intraparticle porosity and the micritic matrix; (D) Thin-section observation under transmitted and fluorescence light (respectively left and right), highlighting the intraparticle porosity; (E) Back-Scattered Electron SEM-micrograph of a thin-section to visualize the intraparticle microporosity and its connection to matrix porosity. Also note the clays (darker) dispersed in the matrix.

Figure 11: Typical NMR T_2 distribution and MICP pore throat radii sizes distribution for 5 lithotypes: micritic, grainy, cemented, argillaceous and silicified chalks.

Figure 12: Schematic representation of the microtexture of (A) micritic chalk: highly porous chalk – about 40% porosity. (B) Cemented chalk: authigenic calcite crystals in the matrix and calcite cement overgrowths on grains (in dark grey). Reduced porosity (about 18%) and pore throat size (and thus also permeability). (C) Argillaceous chalk: clay flakes (in black) floating in the matrix and around micritic grains, reduce porosity (to 25% here), but mostly reduce permeability by reducing the pore throat sizes.

Figure 13: Distribution of the different chalk lithotypes on a ternary diagram of porosity type (chalk-nanoporosity, chalk-microporosity, chalk-mesoporosity) based on porosity versus NMR- T_2 distribution.

Figure 14: (A) $T_2(lm)^2 \phi^4$ versus permeability plot providing gradients used to calculate the constant 'a' used in the SDR model (B) Relationship between NMR-estimated permeability KSDR and actual measured core permeability.

Table 1: Geographic and stratigraphic location of the samples. References from studies which performed geological loggings on the studied outcrops are: (1) Amédéo and Robaszynski, 2000 (2) Amédéo and Robaszynski, 2001 (3) Boulvain and Pinguot, 2012 (4) Bristow et al., 1997 (5) Gale et al., 2005 (6) Gräfe, 1999 (7) Juignet and Breton, 1974 (8) Kennedy, 1969 (9) Lasseur et al., 2009 (10) Marlière, 1949 (11) Mitchell, 1994 (12) Mortimore and Pomerol, 1997 (13) Mortimore, 2011 (14) Robaszynski and Amédéo, 1986 (15) Robaszynski et al., 1998 (16) Whithman, 1993.

Chalk lithotype	Sample Name	Location	Latitude	Longitude	Stratigraphy	Formation name	References
Micritic	OF/CH01	Harmignies Quarry. BE	50.420643	4.0244404	Middle Campanian	Obourg	3, 10
	OF/CH02	Harmignies Quarry. BE	50.420643	4.0244404	Middle Campanian	Nouvelles	3, 10
	OF/CH04	Harmignies Quarry. BE	50.420643	4.0244404	Upper Campanian	Spiennes	3, 10
	OF/CO01	Coquelles Quarry. Boulonnais. FR	50.930955	1.774346	Santonian	Caffier	1, 14
	OF/NH03	Newhaven. Sussex. UK	50.784762	0.023071	Campanian	Newhaven	4, 8, 12, 13
	OF/RA01	Ramsgate. Kent. UK	51.373740	1.448914	Santonian	Seaford	4, 8, 12, 13
Grainy	OF/BG01	Birling Gap. Sussex. UK	50.744115	0.198285	Turonian	New-Pit	4, 8, 12, 13
	OF/CM02	Mimoyecques Quarry. Boulonnais. FR	50.852647	1.759135	Turonian	Guét	1
	OF/CM06	Mimoyecques Quarry. Boulonnais. FR	50.852647	1.759135	Turonian	Guét	1
	OF/ETR33	Etretat. Upper Normandy. FR	49.712103	0.205394	Coniacian	Saint Pierre en Port	7, 9, 12, 15
	OF/SS01	Seven Sisters, Sussex, UK	50.748322	0.187536	Coniacian	Chalk Mudstone	4, 8, 12, 13
Cemented	OF/CB14	Cap Blanc Nez. Boulonnais. FR	50.936655.	1.726030	Upper Cenomanian	Escalles	2, 6, 14
	OF/FA15	Flamborough Head. Yorkshire. UK	54.103918	-0.131857	Santonian	Flamborough	10, 16
	OF/FA39B	Flamborough Head. Yorkshire. UK	54.107806	-0.090544	Santonian	Flamborough	10, 16
	OF/FH11	Flamborough Head. Yorkshire. UK	54.105572	-0.099754	Santonian	Flamborough	10, 16
	OF/SC01	Saint Martin en Campagne. Up. Normandy. FR	49.968511.	1.195942	Turonian	Tilleul	7, 9, 12, 15
Marl-seam	OF/CB13	Cap Blanc Nez. Boulonnais. FR	50.936655.	1.726030	Upper Cenomanian	Escalles	2, 6, 14
	OF/CB16	Cap Blanc Nez. Boulonnais. FR	50.937992.	1.729152	Base Turonian	Grand Nez	2, 6, 14
	OF/EA02	Eastbourne. Sussex. UK	50.759582	0.284676	Base Turonian	Hollywell Nodular	4, 5, 8, 12, 13
	OF/ETR21	Senneville. Upper Normandy. FR	49.784092	0.423037	Middle Turonian	Senneville	7, 9, 12, 15
	OF/ETR47	Senneville. Upper Normandy. FR	49.784092	0.423037	Middle Turonian	Senneville	7, 9, 12, 15
	OF/SC02	Saint Martin en Campagne. Up. Normandy. FR	49.968511.	1.195942	Turonian	Tilleul	7, 9, 12, 15
	OF/SC03	Saint Martin en Campagne. Up. Normandy. FR	49.968511.	1.195942	Turonian	Tilleul	7, 9, 12, 15
Argillaceous	OF/CB02	Cap Blanc Nez. Boulonnais. FR	50.902975.	1.679754	Lower Cenomanian	Strouanne	2, 6, 14
	OF/CB04	Cap Blanc Nez. Boulonnais. FR	50.907931.	1.684760	Lower Cenomanian	Strouanne	2, 6, 14
	OF/CB06	Cap Blanc Nez. Boulonnais. FR	50.907931.	1.684760	Lower Cenomanian	Strouanne	2, 6, 14
	OF/CB07	Cap Blanc Nez. Boulonnais. FR	50.910183.	1.686753	Lower Cenomanian	Petit Blanc-Nez	2, 6, 14
	OF/CB09	Cap Blanc Nez. Boulonnais. FR	50.926157.	1.707652	Mid Cenomanian	Petit Blanc-Nez	2, 6, 14
	OF/CB10	Cap Blanc Nez. Boulonnais. FR	50.926157.	1.707652	Mid Cenomanian	Petit Blanc-Nez	2, 6, 14
	OF/CB11	Cap Blanc Nez. Boulonnais. FR	50.926157.	1.707652	Mid Cenomanian	Cran	2, 6, 14
	OF/CB23	Cap Blanc Nez. Boulonnais. FR	50.911766.	1.688472	Lower Cenomanian	Petit Blanc-Nez	2, 6, 14
	OF/CB24	Cap Blanc Nez. Boulonnais. FR	50.911766.	1.688472	Lower Cenomanian	Petit Blanc-Nez	2, 6, 14
	OF/CB25	Cap Blanc Nez. Boulonnais. FR	50.911766.	1.688472	Lower Cenomanian	Petit Blanc-Nez	2, 6, 14
	OF/EA01	Eastbourne. Sussex. UK	50.759582	0.284676	Cenomanian	Zig-Zag	4, 5, 8, 12, 13
Silicified	OF/BR01	Bruneval. Upper Normandy. FR	49.637.650	0.148597	Cenomanian	Glauconieuse	7, 9, 12, 15

Table 2: Petrographic characteristics of the 35 chalk samples

Chalk lithotype	Sample Name	Macroscopic facies	Grains (%)	Microscopic facies	Insoluble Residue (%)	Diagenesis Index (a.u.)
Micritic	OF/CH01	Chalk Mudstone	1.9	Forams-rich mudstone	0.8	1
	OF/CH02	Chalk Mudstone	1.6	Forams-rich mudstone	0.6	0.5
	OF/CH04	Chalk Mudstone	2.1	Forams-rich mudstone	0.8	0.5
	OF/CO01	Chalk Mudstone	6	Forams-rich mudstone	0.9	1
	OF/NH03	Massive Chalk Mudstone	-	-	0.7	2.5
	OF/RA01	Chalk Mudstone	-	-	0.8	1.5
Grainy	OF/BG01	Massive Chalk Mudstone	-	-	0.4	1
	OF/CM02	Massive Chalk Mudstone	40.1	Bioclastic forams-rich packstone	1.8	1
	OF/CM06	Massive Chalk Mudstone	32.5	Bioclastic forams-rich packstone	0.9	1
	OF/ETR33	Chalk Grainstone	42.8	Bioclastic packstone	0.3	2
	OF/SS01	Massive Chalk Mudstone	-	-	0.5	1.5
Cemented	OF/CB14	Massive Chalk Mudstone	8.3	Calcsphere-rich mudstone	5.9	6.5
	OF/FA15	Massive Chalk Mudstone	4.6	Calcsphere-rich mudstone	3.3	7
	OF/FA39B	Massive Veined Chalk Mudstone	5.7	Calcsphere-rich mudstone	3.5	7.5
	OF/FH11	Massive Chalk Mudstone	5.4	Calcsphere-rich mudstone	3.2	6.5
	OF/SC01	Laminated Chalk Mudstone	-	-	4.3	6
Marl-seam	OF/CB13	Burrowed Laminated Argillaceous Chalk Mudstone	6.3	Forams and sponge spicule-rich mudstone	14.3	3
	OF/CB16	Pebbly Argillaceous Chalk Mudstone	-	Nodular- bioclastic wackestone	7.6	4.5
	OF/EA02	Pebbly Argillaceous Chalk Mudstone	14.1	Nodular- Calcsphere-rich wackestone	5.7	5
	OF/ETR21	Finely Laminated Chalk Mudstone	7.6	Bioclasts and forams-rich mudstone	3.8	1
	OF/ETR47	Burrowed Laminated Argillaceous Chalk Mudstone	16.3	Bioclasts and forams-rich wackestone	2.7	1.5
	OF/SC02	Burrowed Laminated Argillaceous Chalk Mudstone	12	Nodular mudstone	5.7	3.5
	OF/SC03	Massive Chalk Mudstone	-	-	5.4	3
Argillaceous	OF/CB02	Glauconitic Chalk	7.7	Bioclastic mudstone	24.9	3
	OF/CB04	Argillaceous Massive Chalk Mudstone	12.8	Bioclasts and calcsphere-rich wackestone	20.8	4
	OF/CB06	Argillaceous Chalk Mudstone	16.5	Bioclastic wackestone	23.0	3.5
	OF/CB07	Massive Chalk Mudstone	7.9	Calcsphere-rich mudstone	17.5	5.5
	OF/CB09	Burrowed Massive Argillaceous Chalk Mudstone	11.4	Bioclastic wackestone	26.2	4
	OF/CB10	Burrowed Massive Chalk Mudstone	12.9	Calcsphere-rich wackestone	9.7	6
	OF/CB11	Burrowed Argillaceous Chalk Mudstone	17.2	Bioclastic and forams-rich wackestone	29.0	3.5
	OF/CB23	Argillaceous Chalk Mudstone	-	-	17.8	5
	OF/CB24	Massive Argillaceous chalk Mudstone	-	-	27.0	3.5
	OF/CB25	Massive Argillaceous chalk Mudstone	-	-	22.0	4
	OF/EA01	Burrowed Massive Argillaceous Chalk Mudstone	13.6	Calcsphere-rich wackestone	9.8	5
Silicified	OF/BR01	Silicified Chalk	-	Silicified mudstone	76.8	8

Table 3: Petrophysical results on the 35 selected chalk samples

Chalk lithotype	Samples Name	Ambient Porosity (%)	Klinkenberg Permeability (mD)	Grain density (g/cm ³)	T _{2m} (ms)	Average Pore throat radius (nm)	Specific surface area (m ² /g)
Micritic	OF/CH01	41.9	3.18	2.70	57.8	249	2.17
	OF/CH02	44.6	4.48	2.71	44.1	270	2.2
	OF/CH04	42.4	2.85	2.71	37.5	194	-
	OF/CO01	42.9	5.83	2.70	50.7	-	-
	OF/NH03	28.5	1.84	2.71	45.0	-	-
	OF/RA01	45.9	5.95	2.70	47.3	310	-
Grainy	OF/BG01	37.8	3.09	2.68	90.0	-	-
	OF/CM02	39.7	3.67	2.70	62.9	302	-
	OF/CM06	43.6	4.32	2.71	86.2	-	-
	OF/ETR33	33.8	13.30	2.70	88.2	540	0.8
	OF/SS01	40.2	3.10	2.68	80.5	296	-
Cemented	OF/CB14	27.1	0.43	2.70	16.2	113	2.7
	OF/FA15	17.6	0.10	2.71	15.3	-	-
	OF/FA39	16.2	0.14	2.71	15.7	114	1.9
	OF/FA11	19.3	0.16	2.71	16.3	-	-
	OF/SC01	23.8	0.44	2.70	17.4	137	-
Marl-seam	OF/CB13	31.4	0.30	2.68	8.6	61	8.6
	OF/CB16	20.7	0.19	2.71	12.0	-	9
	OF/EA02	14.5	0.25	2.70	3.9	50	-
	OF/ETR21	36.4	1.32	2.70	31.7	213	-
	OF/ETR47	40.4	2.68	2.68	29.9	-	-
	OF/SC02	33.0	0.48	2.68	10.4	103	-
	OF/SC03	25.4	0.40	2.69	21.0	-	-
Argillaceous	OF/CB02	21.6	0.10	2.69	3.5	43	-
	OF/CB04	18.8	0.06	2.69	3.6	-	-
	OF/CB06	23.4	0.11	2.68	4.3	54	-
	OF/CB07	21.4	0.13	2.70	10.3	-	-
	OF/CB09	22.1	0.08	2.68	5.1	48	-
	OF/CB10	18.9	0.10	2.70	5.9	61	-
	OF/CB11	23.8	0.10	2.69	7.0	38	11.23
	OF/CB23	21.0	0.06	2.68	4.0	-	-
	OF/CB24	18.2	0.08	2.67	5.7	-	-
	OF/CB25	20.1	0.06	2.69	4.9	-	-
	OF/EA01	14.2	0.06	2.72	3.6	113	-
Silicified	OF/BR01	26.4	0.04	2.45	17.6	12	-

Highlights

- Low-field NMR T_2 distributions allow to differentiate 5 chalk lithotypes
- Each lithotype displays distinct petrographical and petrophysical properties
- Main permeability-reducing factors in chalks: non-carbonate content and cements
- Bimodal NMR signal of marl-seams is related to intraparticle porosity preservation
- Permeability can be inferred from NMR measurements using the SDR model