The geology of roofing slate

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Abstract: This paper reviews the geological factors linked to the quarrying and quality of roofing slate deposits, and gives recent research results on the loss of fissility by dewatering, and the oxidation of iron sulphides. Mineralogy, resulting from the original protolith composition and the metamorphic grade, controls the colour, brightness and durability of slate tiles. The microtexture of the rock mainly controls the split fitness of slate (fissility). The structure (folding, slaty cleavage development, S_0–S_1 angle, lack of crenulation cleavage and kink-bands, joints and faults) controls the exploitability. Additional factors, such as bed thickness and changes in facies are also important. Both very cold environments, giving frost, and arid conditions, drying the rock enough to remove water, produce a loss of fissility in slate. Ostwald-ripening by dewatering and some collapse of the crystallographic structure of phyllosilicates cause the loss. A mineralogical study of the iron sulphides in some Spanish slate (pyrite, pyrrhotite, marcasite), encompassing crystal habit and size, and their biochemical alteration processes (stain spots), allow the oxidation behaviour of different slates to be predicted.

The mining and processing of slate is an industry with a world production of about one million tonnes per year, roughly valued at €370 million (Euros). The EU is the largest producer, dominated by northwestern Spain. This paper reviews the roofing slate industry and the published technical literature, discusses the main geological features relevant to the extraction of roofing slates, and gives a detailed description of the major slate producing area in Spain, the Truchas Syncline. It also discusses some recent research on the dehydration of slate and the oxidation of minor sulphides, sometimes found in slate products.

The roofing slate industry

The extraction of slate and other fissile rocks for roofing is probably as old as the art of building with stone. Although they used terracotta tiles more usually in their major buildings, the Greeks and Romans also used slate and marble for roofing. For example, slate roof remains of Roman buildings are known in Cumbria and the Cotswolds, United Kingdom (Briggs 1954). During the Dark Ages (around AD 400–1000) the use of slates for roofing became much diminished, timber being widely used in Romanesque buildings. Fire danger (thatch roofs were forbidden in London in 1212), however, gradually encouraged the use of safer slate and lead in later centuries. Historical centres of many European cities include Medieval and Renaissance buildings covered with slate.

A well developed international trade in roofing slate commenced in the late 19th Century, as European construction fashion and materials were exported to the New World. French, German and Welsh quarries increased their activity and began to export slates. For example, in many major buildings of that period in Buenos Aires, Argentina, Italian Carrara marble and French slate were used. Much Welsh slate was exported to the Americas and other parts of the British Empire. This production and export pattern continued through the first half of the 20th century. However, a rapid change in the international production and market of roofing slate took place in the 1970s (Lombardero & Regueiro 1992). Traditional producers such as France (then the main producer), Germany and the United Kingdom, with high costs of mining (some underground), lost their market share. This was caused by a rapid increase in exploitation of Spanish deposits, many of which were virgin, and as a result, the slate was more easily and more cheaply available. Today, Spain is the principal producer of slate tiles, accounting for nearly 80% of the 900 000–1 000 000 tonnes annual total world production. National companies now control production in Spain and a very important part of the international market.

Technical literature on slate

There are numerous publications on roofing slate quarries. For example: Germany (Pfeiffer 1955, 1956; Steinbach *et al.* 1967; Wagner 1990), Norway (Kolderup 1959), United Kingdom (Crockett 1975), North America (Bell 1863; Evans & Marr 1988; Conrad & Vanecek 1995). The quality and exploitability of slate are discussed by Le Corre (1968, 1969, 1970). The results of extensive exploration and research into the slate deposits of Spain, much carried out by the Spanish Geological Survey (IGME) during the 1980s, are published in Ruiz-Garcia (1977), Barros *et al.* (1985), Lombardero (1988, 1994), Lombardero & Quereda (1992), Barros (1994), Toyos *et al.* (1994), López-Jimeno (1995), Lombardero & Toyos (1995) and García-Guinea *et al.* (1997). Spanish researchers have established the geological factors controlling the quality of roofing slate in Spain, and by working closely with the slate-producing companies, have created a prospecting methodology during the 1980s and 1990s. This research also helped to provide explanations to the quarrymen of former 'unpredictable' factors arising during exploitation.

Other specific research on slate properties includes Manning (1975) and Shayan & Lancucky (1984). Other work has been reported on the influence of dewatering on the split attitude of the slate and on the biochemical process of oxidizing iron sulphides of the slate (García-Guinea *et al.* 1998, 2000). These two aspects have important industrial relevance, because they strongly affect the yield and quality of slate roofing tiles.

Geological features

The European Committee of Standardization (CEN 1997) gives the following definitions:

Slate: A fine-grained very low- to low-grade metamorphic rock possessing a well-developed fissility parallel to the planes of slaty cleavage.
Roofing slate: Rocks that are easily split into thin sheets along planes of slaty cleavage, caused by very low- to low-grade metamorphism due to tectonic compression. Phyllosilicates are

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Table 1. *Mineralogy of roofing slates*

Minerals	Amount	Genetic Type
White mica mica (illite–muscovite ± phengitic (Na, K)-mica, ± paragonite)	40–60%	Mainly metamorphic but some large muscovite grains are sedimentary (detrital)
Chlorite and/or chlorite–mica stacks	15–20%	Metamorphic and some detrital or diagenetic
Fine-grained quartz	22–25%	Detrital but elongated and recrystallized by metamorphism
Chloritoid	0–10%	Metamorphic
Albite, graphitic matter,	absent or minor	Metamorphic
Rutile, ilmenite, zircon, tourmaline, apatite, K-feldspar, small lithic and fossil fragments	absent or minor	Sedimentary (detrital)
Carbonates	0–10%	Sedimentary
Fe-, Cu- and Zn-sulphides	absent or minor	Diagenetic or metamorphic

the predominant and most important components. These are mixed with fine-grained quartz and larger or smaller quantity of clay minerals, depending on the metamorphic grade.

The most conspicuous feature of slate is, obviously, that the rock has slaty cleavage. This is also defined by CEN (1997):

Slaty cleavage: A variety of foliation, typical of fine-grained metamorphic rocks such as slates, consisting in continuous and homogeneous preferred orientation of mineral grains, especially the platy crystals of mica that show a plane texture visible under polarizing microscope.

Slate is usually made up of a mix of sedimentary, diagenetic and metamorphic minerals (García-Guinea *et al.* 1997). These are listed in Table 1. In one quarry in Spain, chlorite-rich slates are reported to be of better quality than those with less chlorite (Blanco *et al.* 1988). The green colour of some slates is due to a high percentage of Fe-rich chlorite. Graphitic matter produces grey and black colour slates. Those with less quantity of graphitic matter and more quartz are usually light grey. Carbonates are rapidly leached in acid environments and cause white spots on the slate surface. Slates with large amounts of sulphides (pyrite, pyrrhotite, marcasite, chalcopyrite, occasionally sphalerite) are generally of lower quality (Ruíz-García 1977; Lombardero & Toyos 1995; García-Guinea *et al.* 1997). The bio-chemical hydroxylation of marcasite and pyrrhotite is very fast, and is accompanied by volume changes and acid generation. This results in stain spots and ribbons on the roof. Pyrite is more resistant, decaying only in acid environments (i.e. in response to acid rain or peat cover, or in association with pyrrhotite).

The best roofing slate is quarried from massive meta-pelitic formations that have experienced low-grade regional metamorphism under low greenschist facies, i.e. chlorite facies (temperature 300–400°C, pressure 2–3 kbar). Most have white mica (illite) crystallinity values (Kubler index) of 0.15–0.25 $\Delta°2\theta$ and are therefore epizonal rather than anchizonal. The presence of paragonite and/or (Na, K)-mica leads to anomalously high index values, and therefore indicates apparently slightly lower grade (Roberts *et al.* 1990).

The authors have never observed biotite in good quality roofing slate. Moreover, in the areas where progressive regional metamorphism can be mapped, the biotite + isograd marks the highest metamorphic boundary of exploitable slate deposits. Despite this, exploitable slate from Virginia (USA) contains minor amounts of biotite (Evans & Marr 1988).

Thermal metamorphism, induced by plutonic bodies younger than regional metamorphism or other thermal events, produces recrystallization and formation of new minerals (andalusite, biotite, garnet etc.) that weld the fissility planes, impeding good splitting of the slate. Both texture and microstructure are very important factors in fissility or split fitness of the slate, controlling the yield of the rock when processed into tiles. The main considerations are:

General texture: The rock should be lepidoblastic with a primary continuous slaty cleavage (S_1), generated by recrystallization under strain during the first main tectonic episode of regional metamorphism (Figure 1A).

Average grain size. Quartz grains are usually less than 75 µm (measured perpendicular to foliation). Slates can be classified as fine grained: <30 µm, usually very good fissility; medium grained: 30–50 µm, good fissility; coarse grained: >50 µm, usually medium to poor fissility.

Homogeneity of grain size. This is a critical factor. Fine-grained slates with a few porphyroblastic chlorite or chloritoid crystals of 100–200 µm can have poorer fissility than an homogeneous coarse grained slate without porphyroblasts (Figure 1B), big detrital grains of quartz or muscovite, lithic fragments, fossil remains, ribbons of quartz, etc. (Lombardero 1994; Lombardero & Toyos 1995; García-Guinea *et al.* 1997).

Layering. Microlayering (S_0) produces parallel strips in slate with slight differences in quartz percentage and grain size, that break the textural homogeneity and decrease the fissility by cleavage refraction, especially if the angle S_0–$S_1 > 10°$ (Figure 1C).

Lineation. Termed *hebra* or *freba* (string) by Spanish quarrymen, is the L_{0-1} or intersection S_0–S_1. It is easily visible on the fissility (cleavage) plane if the slate bed is not massive and microlayering appears. It affects the slate processing (when it is sawn).

Longrain. Penetrative mineral lineation on the cleavage plane (X-Y plane of the strain ellipsoid), nearly parallel to the Y axis. It is termed *grain* by Welsh quarrymen, *hilo* (thread) in Spain and *downdip lineation* (Hobbs *et al.* 1976), and is also important in the processing stage of slate. Usually, orientated rutile and other acicular minerals define this lineation (Le Corre, 1968), that may or may not be parallel to L_{0-1}. Potential for confusion exists because US quarrymen refer to L_{0-1} as grain instead of *longrain*.

Secondary foliations. The presence of one (or more) foliation (S_2) younger than the S_1, such as a crenulation cleavage, causes at best, irregular fissility surfaces that do not allow flat slate tiles to be produced or, at worst, provides discrete terminations of S_1. If the angle between S_1 and S_2 is low, a crenulation cleavage is usually poorly developed. Instead, characteristic, smooth, glossy surfaces appear, termed *pizarra quemada* (burnt slate) that may lead to low-angled wedges of slate, instead of parallel-faced sheets. The presence of small-scale kink-bands, mineral veins, such as quartz veins or quartz-chlorite veins, causes similar problems.

The meso- and macro-structure is another factor that controls the exploitability of slate deposits. The main points to take into account are:

Bed thickness: Beds of less than 10 m thickness are not usually exploited. Nevertheless, in some quarries metre-scale slate layers inter-bedded with sterile lithologies are quarried, if the inter-bedded formation is at least 100 m thick and the slate layers are over a half of the total. Bed thickness may not be continuous, due to folding.

Folding. Metapelitic slate formations are intensively folded, generally with more than 50% shortening, thus folds are of similar type (Figure 1D), with low angles between limbs, and cleavage nearly parallel to the axial plane. This fold style produces hinge thickening in slate beds, reaching a 6 : 1 ratio (hinge thickness: limb thickness), which sometimes allows the exploitation of thin slate layers only at the hinges. A few Spanish quarries are in this structural position. The structural setting must be studied, especially if the slate bed is not massive (i.e. it has thin silty or sandy laminations), as if the angle S_0–S_1 is higher in the hinge than in the limbs, the extraction-processing yield will probably be lower in the hinge than in the limbs. Also, tiles will have a marked L_{0-1} lineation on their surface.

Discontinuities. Joints, faults, large kink-bands and veins are all discontinuities in the slate rock bodies. These are also very important factors in the assessment and mining of slate (Toyos *et al.* 1994; Lombardero & Toyos 1995; García-Guinea *et al.* 1997). A careful measurement of the discontinuities in the field together with a statistical treatment of data allow the average-sized block and the percentage of blocks bigger than a specified minimum existing in an outcrop or quarry to be ascertained. The minimum-sized block depends upon the quality of the slate and the efficiency of the extraction-process: the better quality and more efficient the process, the smaller the volume of the minimum block size.

The Truchas Syncline in NW Spain

The Truchas Syncline is currently the major roofing slate producing area of the world. Since 1980, the IGME, universities and private companies have intensively studied this megastructure. Therefore, there is a good geological knowledge of the area (Figs 2 & 3). Like all the Spanish (and French and Portuguese) slate producing areas, it belongs to the West European Hercynian Range, termed Iberian Massif in Spain and Portugal. It originated by continental collision in Carboniferous to Lower Permian times. Three main tectonic phases have been recognized. The first, D_1, led to the development of all-scale similar folding and the primary cleavage S_1. D_2 caused large-scale thrusts, which carried internal zones over external zones. The third phase, D_3, produced major folds, commonly with greater wavelength. D_3 also caused much more open folds (high inter-limbs angle) than D_1, and a number of minor structures such as crenulation cleavage, kink-bands, normal and deep-direction faults. This polyphase deformation was accompanied by polyphase regional metamorphism and plutonic intrusion, especially in the last deformation stage. The Truchas Syncline is a major D_3 fold, developed on a tightly folded but, in general, normal limb of a D_1 fold. All inverse limbs observed belong to D_1 folds. Primary cleavage trends WSW–ESE, dipping gently (south limb) to 50° (north limb) to the south.

The stratigraphic sequence is marine and comes from Lower Ordovician (Tremadocian) slates to Silurian ampelites and quartzites. Productive units are the *Pizarras de Luarca* (Luarca slates) formation (Llanvirnian–Llandeilian), mainly composed of massive slate and some metavolcanics, and Casaio and Rozadais formations (Caradocian–Ashgilian), composed of slates, (some containing dropstones of Hirnantian Glaciation origin), slates with sandy or silty laminations, quartzites and a few small reef limestone bodies. Exploitable units of equivalent age and facies are found in the Armorican Massif in France (*schistes á Calymene* = *Pizarras de Luarca* and *Schistes á Trinucleus* = *Casaio* + *Rozadais* formations) and Portugal (*Ardesia de Valongo*). The main quarries of good quality roofing slate are in Ordovician formations in a peri-Gondwanic marine sedimentological environment in Western Europe, North America and even in Argentina (Lombardero & Reile 1997).

The principal quarries are in the South limb of the Truchas Syncline, although the structural and stratigraphic location varies. In San Pedro de Trones they are on the Rozadais Formation on the normal limb and, partly, the hinge zone of a hectometric overturned fold, with gently dipping (8°–15°) cleavage. The Penedo Rayado and Os Foyos quarry zones are in the same formation on a normal limb, the cleavage dipping some 20°. Mormeau and Juanita-Los Molinos quarries are in a 15 m thick slate bed of the Casaio Formation. They are in the hinge zone of a syncline and anticline, respectively, where exploitable rock massifs approach dozens of metres in thickness. The north limb of the syncline has a higher dip angle than S_1, and is more affected by S_3 crenulation, faults and kink-bands. On this limb quarries are scarce, but there are a few that are very productive.

About 60% of the good quality roofing slate marketed in the world is produced in the Truchas Syncline. Products include a wide range of black, dark grey and grey tiles, in many formats and sizes. Around 4000 people work in the quarries and processing factories, which is the main economic activity in the region.

Recent research

Slate dehydration

Roofing slate loses a great amount of its fissility when dry. This is well known by quarrymen. They keep the blocks of rock wet in dry weather, and they keep sub-blocks sawn from these large blocks wet until they are split. Dried slate does not recover its fissility even when submerged in water for a long time. Keeping slate from drying out increases the cost of processing, and for mechanised processing, a procedure which does not require the slate to be kept wet would be beneficial. Understanding the mechanism of drying of slate will aid the development of new processing equipment.

The dewatering process has been studied using thermal X-ray diffraction (TXRD) in a simultaneous optical stimulated (SOS) X-ray diffraction device developed in the laboratories of the Spanish Museum of Natural Sciences, in Madrid. During heating, in the 2θ interval 7.5–9.5°, phyllosilicate diffraction peaks move and the intensity of the diffraction changes (Fig. 4), especially when the temperature is near 100°C. Large changes in the illite–chlorite TXRD 10Å peak have been detected by isothermal scanning (i.e. 95°C) of TXRD under thermo-photonic stimulation and electronic temperature control. These large anomalies of the illite crystallinity peak, working with aliquot samples of slate, prevent the common petrologic use of the Kubler index using the slaty cleavage of micro-mono-blocks.

The irreversible drying could be caused by the closure of porosity following an Ostwald Ripening type process of illite through the slaty cleavage interfaces, along with the leaking out of alkali-rich aqueous solutions. The random drying of roofing slate produces swelling of layers and crystallite coalescence.

Oxidation of sulphides in slate

The mineralogical composition of roofing slate includes small amounts of iron and other metal sulphides (Fig. 1E). If these

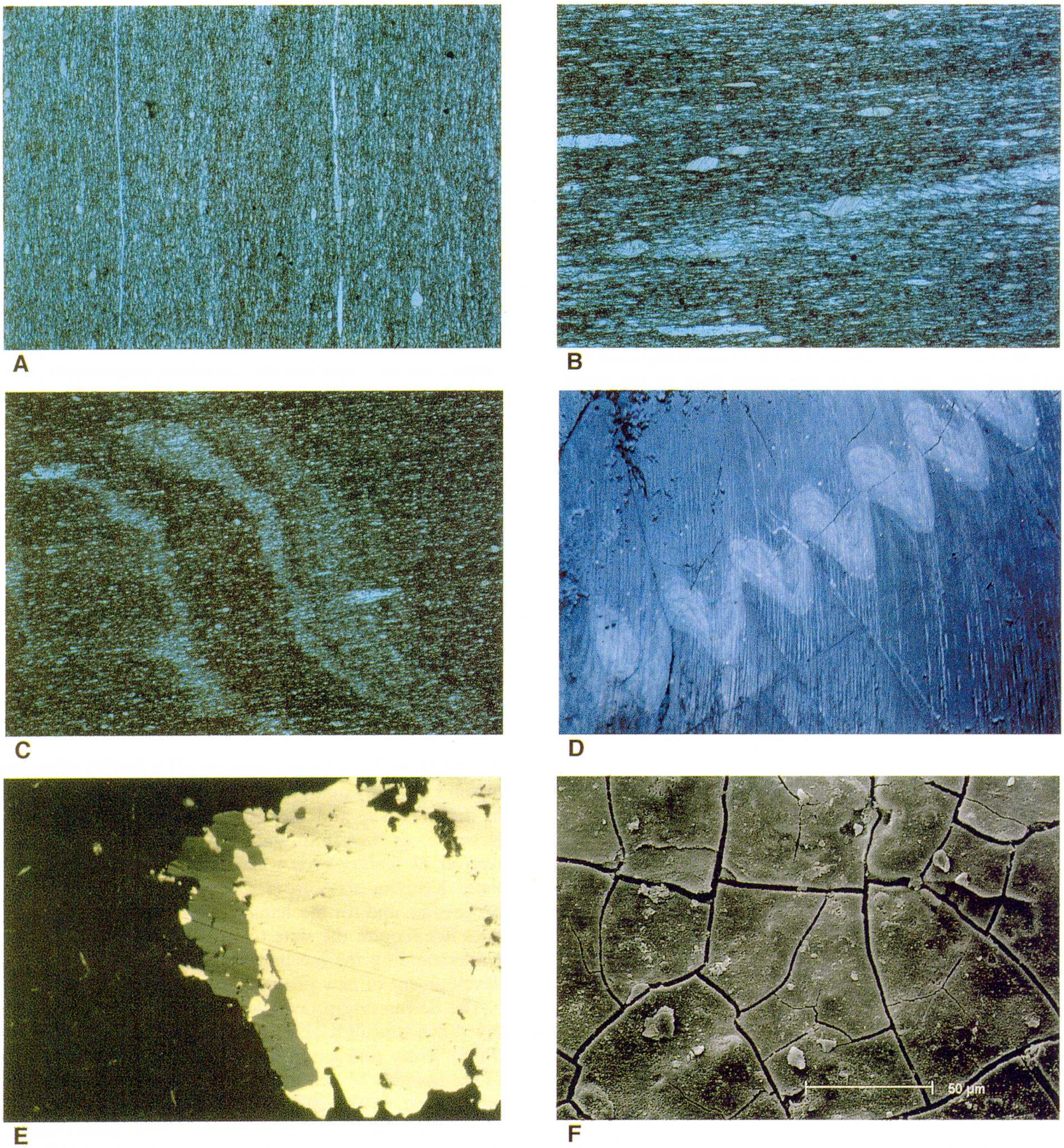

Fig. 1. (A) Photomicrograph of a fine grained roofing slate with a continuous, well developed slaty cleavage. Plane polarized light. Field of view 1.4 mm. (B) Photomicrograph of a coarse grained roofing slate with remains of microlayering (stratification) and some microporphyroblasts. Plane polarized light. Field of view 3.5 mm. (C) Photomicrograph of a fine grained roofing slate with microlayering. Plane polarized light. Field of view 1.4 mm. (D) Centimetre scale folds developed in a slate formation (Corporales, León, Spain). Field of view approx. 300 mm. (E) Photomicrograph (reflected light, plane polarised) of pyrite and pyrrhotite crystals in roofing slate. Field of view 3.5 mm. (F) SEM image of ferric hydroxide coating developed by chemical attack on pyrrhotite. Field of view 0.01 mm.

sulphides oxidize during service, then the slate tile becomes stained. In order of abundance, the sulphides in Spanish roofing slate are pyrite, pyrrhotite, chalcopyrite, sphalerite and marcasite (identified by electron microprobe and scanning electron microscopy). The sulphides in some slate tiles decay rapidly by weathering when the tiles are installed, or even beforehand, during outdoor storage on pallets. This oxidation is well known by producers, who classify slates into two main categories: those with oxidizable ‘pyrite’, and those without. The price of slate tiles having oxidizable iron sulphides is about 30% less than the price of first quality tiles. Thus, understanding the nature of the sulphides and their oxidation is of commercial importance to the industry.

The oxidation involves many processes. The most important may be the biological processes caused by *Thiobacillus* and *Leptobacillus ferrooxidans* (Schippers *et al.* 1996; Sasaki 1997; Barcelar-Nicolau & Barrie 1999), which may attack the iron sulphides in an indirect way (increasing the pH) or in a direct way by oxidizing the mineral surface. Other factors affect the kinetics of the oxidation. These include the habit of the iron

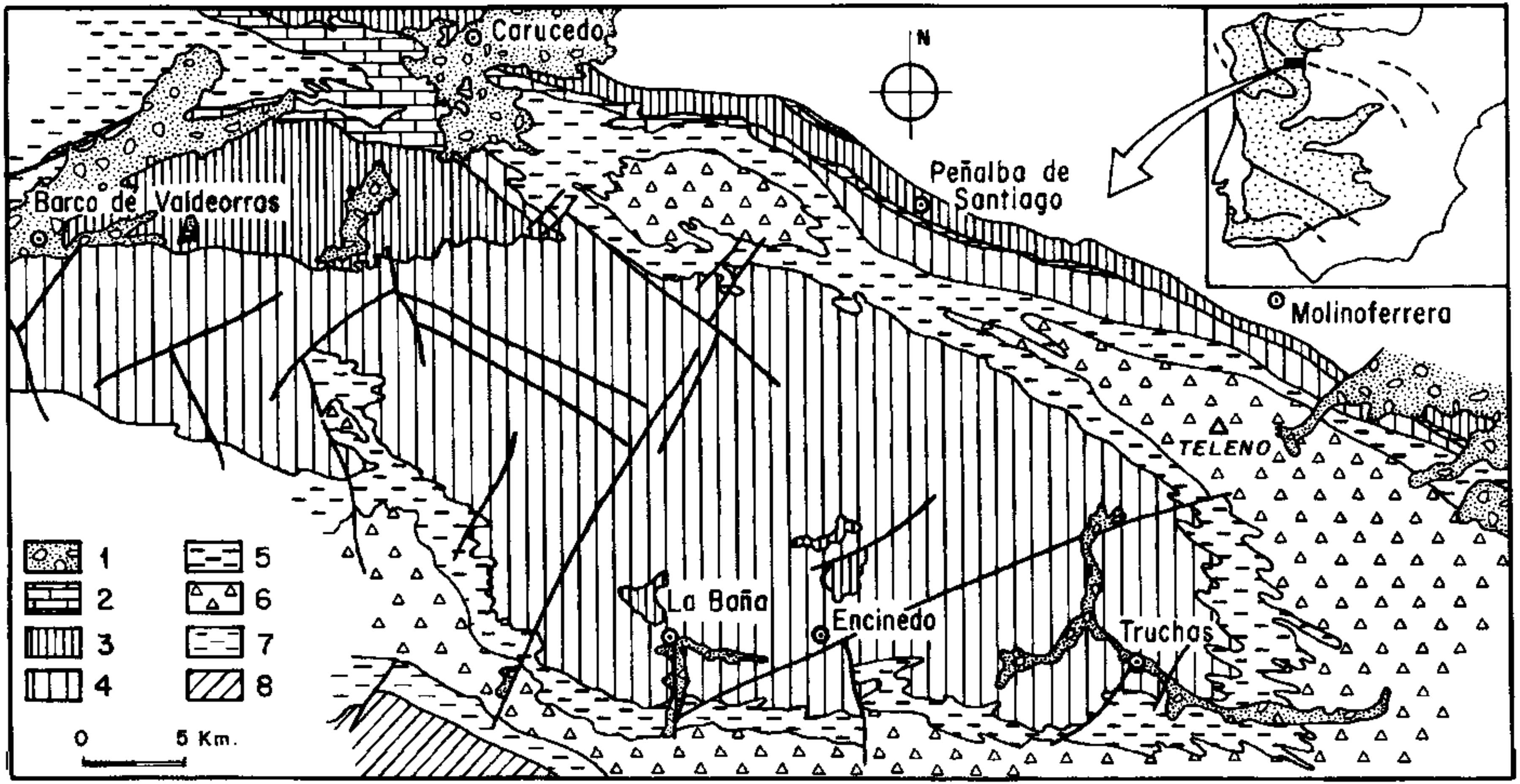

Fig. 2. Geological sketch map of the Truchas Syncline, NW Spain: 1, Tertiary and Quaternary sediments; 2, La Aquiana limestone (Upper Ordovician); 3, Ampelitic slates and quartzites (Silurian); 4, Losadilla + Rozadais + Casaio formations (Upper Ordovician); 5, Luarca Slate formation (Middle Ordovician); 6, Armorican Quartzite formation (Lower Ordovician); 7, Los Montes Slate formation (Lower Ordovician); 8, Ollo de Sapo meta-igneous formation (Upper Cambrian Lower Ordovician).

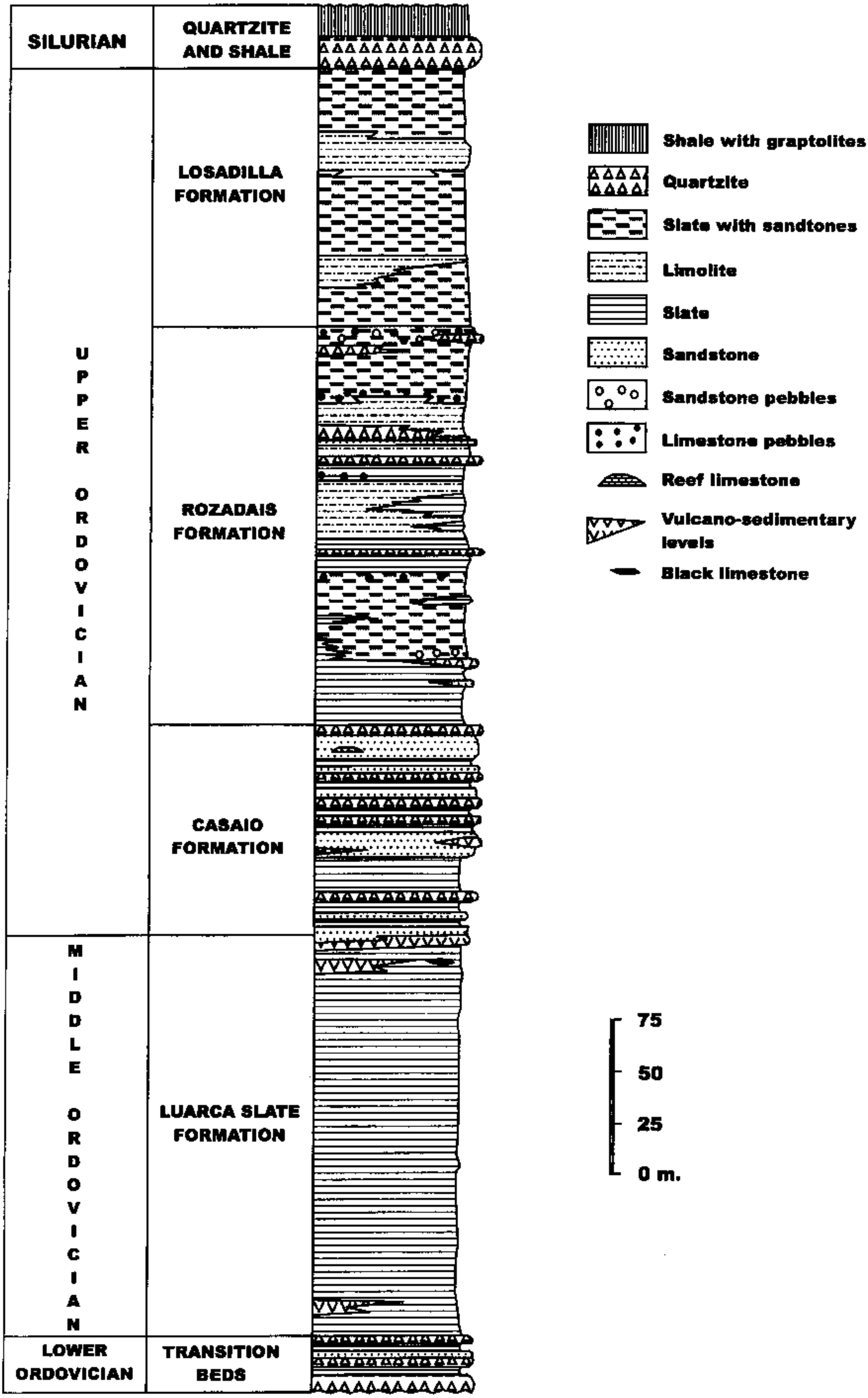

Fig. 3. Stratigraphical section of the South Limb, Truchas Syncline. Limolite, silty slate.

sulphides, the environmental conditions (including temperature, pH and the chemistry of water) and the environment of use of the slate.

Selective mining to avoid slate known to contain sulphides or replacing tiles which show staining are ways to avoid or solve the problem; but, these are expensive solutions and do not adequately tackle the problem. Some current research involves creating a hydroxide coating on the surface of the sulphide (microencapsulation) by chemical attack, following the work of Evangelou (1995) and Zhang & Evangelou (1998). It can be shown that a ferric hydroxide coating can form on the surface of iron sulphides by attacking the tile with various solutions and bacteria incubations (Fig. 1F). The coating is more evident in pyrrhotite than in pyrite, due to the more disordered crystalline structure of pyrrhotite which allows a higher rate of 'free' Fe^{2+}. With further standard tests (UNE-EN 12326–2 Standard) is possible to establish the effectiveness of the microencapsulation. Coating iron sulphides may be an effective and cheap solution, enabling the use of sulphide-bearing slates, but the technology has to be proved further with further laboratory and industrial scale testing.

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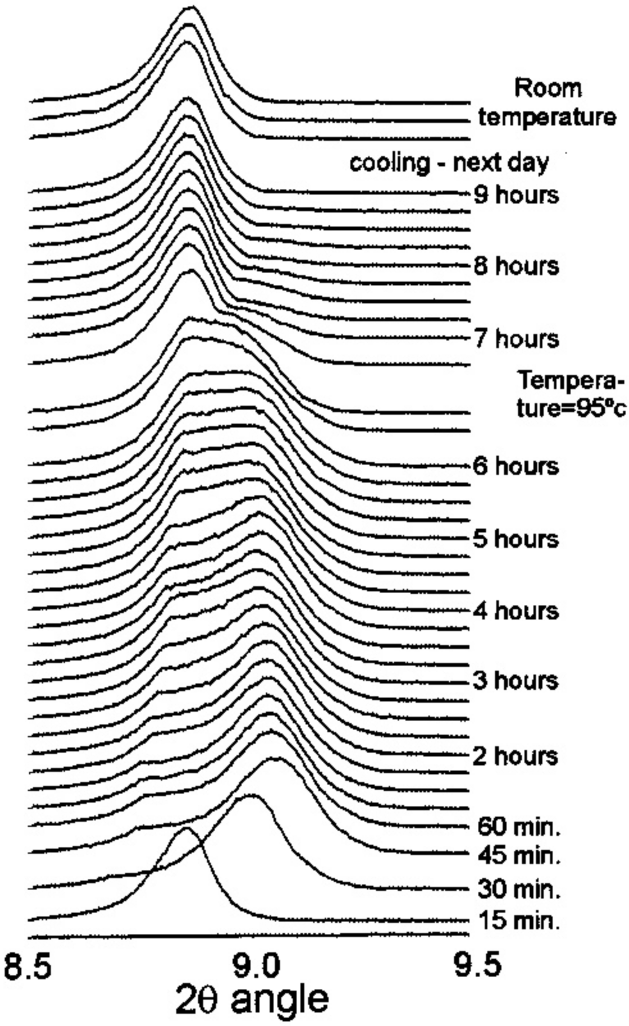

Fig. 4. Migration of (001) illite–chlorite main peak during slate dehydration at 95°C (after García-Guinea *et al.* 2000).

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