Characterization of potential swelling risk of pyritic black shales using MENARD pressuremeter

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ABSTRACT

Black shales are soils and rocks composed of varying amounts of organic matter, clay minerals, carbonates, and sulphides such as pyrite. Toarcian black shales, outcropping in the southern part of Luxembourg, are geotechnically significant due to their potential to swell, causing differential ground heaving and severe structural damage. Unlike classical swelling, which typically involves clay mineral expansion or sulphate phase transitions, the swelling in black shales results from complex pyrite oxidation. When exposed to moisture and oxygen, pyrite (FeS_2) rapidly transforms into secondary minerals such as calcium sulphates and iron oxyhydroxides. These transformations lead to substantial volume increases and pressures that can damage overlying structures. Therefore, geotechnical assessments rely on measuring pyrite content at various depths using laboratory techniques such as X-ray diffraction and differential thermal analysis. These are performed alongside in-situ geotechnical investigations using pressuremeter tests to evaluate swelling potential. Experience with the MENARD pressuremeter has shown that the swelling risk can be effectively inferred from pressuremeter limit pressure (pl_M) values. For black shales with $pl_M > 1.2$ MPa, the risk of swelling is high, and special construction measures are advised to mitigate damage from pyrite alteration. Conversely, in black shales with $pl_M < 1.2$ MPa pyritic swelling only occurs in exceptional conditions. This approach enables a reduction in costly and time-consuming laboratory analyses by inferring the lithological pyrite content and swelling risk directly through pressuremeter results used as well for the common geotechnical design of the foundations. In this paper we discuss the updated laboratory and pressuremeter results from the paper presented 2013 at the ISP6 (Heintz et al. 2013).

RESUME

Les schistes noirs sont des sols et des roches composés de quantités variables de matière organique, de minéraux argileux, de carbonates et de sulfures tels que la pyrite. Les schistes noirs toarciens affleurant dans le sud du Luxembourg, présentent un intérêt géotechnique important en raison de leur potentiel de gonflement, provoquant des soulèvements différentiels du sol et de graves dommages structuraux. Contrairement au gonflement classique, qui implique généralement une expansion des minéraux argileux ou des transitions de phase sulfatée, le gonflement des schistes noirs résulte d'une oxydation complexe de la pyrite. Exposée à l'humidité et à l'oxygène, la pyrite (FeS_2) se transforme rapidement en minéraux secondaires tels que les sulfates de calcium et les oxyhydroxydes de fer. Ces transformations entraînent des augmentations de volume et des pressions substantielles qui peuvent endommager les structures sus-jacentes. Par conséquent, les évaluations géotechniques reposent sur la mesure de la teneur en pyrite à différentes profondeurs à l'aide de techniques de laboratoire telles que la diffraction des rayons X et l'analyse thermique différentielle. Ces évaluations sont réalisées parallèlement à des investigations géotechniques in situ utilisant des essais pressiométriques pour évaluer le potentiel de gonflement. L'expérience avec le pressiomètre MENARD a montré que le risque de gonflement peut être efficacement déduit des valeurs de pression limite pressiométrique (pl_M). Pour les schistes noirs dont la pl_M est supérieure à 1,2 MPa, le risque de gonflement est élevé et des mesures de construction spécifiques sont recommandées pour atténuer les dommages causés par l'altération pyritique. À l'inverse, les schistes noirs dont la pl_M est inférieure à 1,2 MPa présentent un risque de gonflement pyritique négligeable, qui ne survient que dans des conditions exceptionnelles. Cette approche permet de réduire les analyses de laboratoire coûteuses et longues en déduisant directement la teneur en pyrite et le risque de gonflement à partir des résultats des essais pressiométriques in situ, utilisés également pour la conception géotechnique courante des fondations. Dans cet article, nous présentons les résultats actualisés des essais de laboratoire et de pressiomètre, issus de l'article présenté en 2013 à l'ISP6 (Heintz et al. 2013).

Keywords: pyritic black shale, pyritic swell, swell risk as a function of pressuremeter limit pressure threshold values, damage patterns and geotechnical design aspects.

1. Introduction

Fresh black shales are fine-grained thin layered sedimentary rocks composed of abundant organic matter, clay minerals, sulfides (notably pyrite), and carbonate matrix minerals. The observed sulfides record the primary anaerobic deposition conditions. In geotechnical engineering projects these lithologies are known for their swelling potential (Wagner 2013). As a result, these rocks require special attention from the geotechnical engineers due to the significant potential swelling risks. Black shales are responsible for differential heaving and foundation failure due to swelling when exposed to oxygen and moisture. The Fig. 1 reported global distribution of black shale formations in space and time, makes the here reported geotechnical engineering problem also of global interest while its potential consequences are however often underestimated or even neglected, despite the longstanding warning of Stiny (1922) within his then pioneering text book „Technische Geologie“ mentioning „Special care must be taken with clays that are rich in pyrite, because the weathering of pyrite in humid air ... causes strong swelling and creep phenomena under spatial expansion“ (page 546).

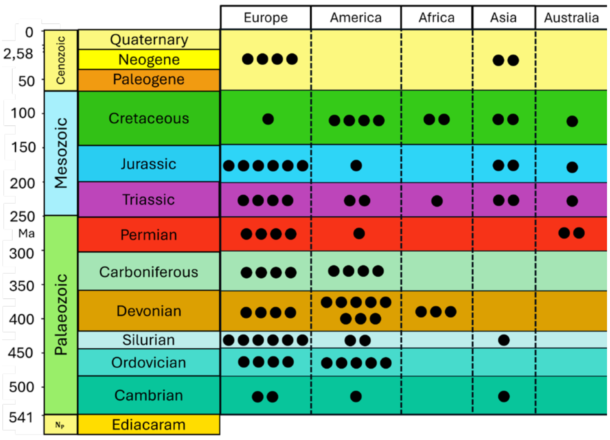

Fig. 1. Distribution of post-precambrian black shale formations in space and time based on Tourtelot (1979)

Unlike to the well-known classical expansive soil damages, where swelling is primarily driven by simple smectite-group clays and/or sulfate mineral transitions, swelling in black shales is a complex mineralogical reaction chain mainly attributed to sulfide cf. pyrite alteration (oxidation). This is a geological ongoing chemical weathering process in near surface black shale formations, but the reaction can also be triggered and/or accelerated by geotechnical construction works. The pyrite alteration reaction leads to the formation of secondary minerals such as gypsum ($CaSO_4 \cdot 2H_2O$), jarosite, and iron oxyhydroxides (e.g., goethite) and a significant volume increase in the rock. Pyrite has a volume of 23.9 mm^3/mol while the product gypsum has a molar volume of 74.7 mm^3/mol (Szmanski, 1989) which represents an approximal volume increase of 310vol.% and corresponds so to the 350vol.% deduced from crystallographic facts by Fasiska et al (1974). The actual in situ increase in volume of the rock formation is however generally much lower as it is intrinsically linked to the density of the pyrite distribution and the associated degree of weathering (see sections 2 and 3).

It should only be briefly mentioned here that geobiological processes (e.g. by sulfide-oxidizing bacteria) accelerate the inorganic reaction equations described in this paper.

Our experience confirms the observation of Tietze (1981), that the resulting crystallization of gypsum in the thin layered strata of black shales is exerting a heave pressure that can exceed 300 kN/m². Geotechnical constructions and geological overlay loads, within sulfide bearing black shale formations, which are below this value are subject to surface uplift due to the swelling lithology.

Due to the fact, that like an increase in volume, heave pressure depends on the density of the pyrite distribution and its alteration status (weathering degree); therefore it can't be simplistically described with reported much higher crystallization pressures. In this context it should be remembered that for example hydrate formation during the conversion from anhydrite ($CaSO_4$) into gypsum ($CaSO_4 \cdot 2H_2O$), generates pressures in the order of 110 MPa (Correns 1981).

In Fig. 2 we included the observation of Tietze (1981) made in similar geological conditions which can be also observed in Luxembourg, which was later implemented as a reference value in Germany within a technical communication for inspection engineers (vpi, 2022).

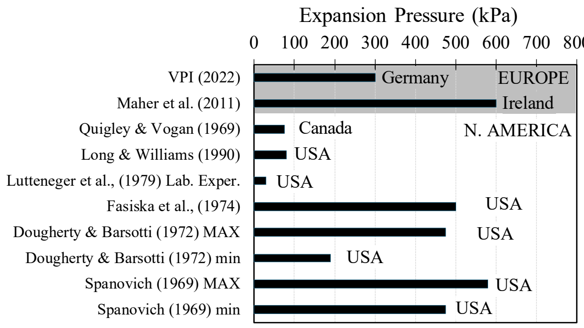

Fig. 2. Comparison of estimated pyrite heave pressures from a literature review (modified from Maher et al 2014 with references therein and updated with new data)

Fig. 2 illustrates that measured values higher than the reference value of 300 kPa are not uncommon, which calls for caution; in contrast lower values could be related to an increased degree of weathering of the studied lithologies, but alteration degrees where unfortunately not reported.

Furthermore, the mineral transformations are not only responsible for a volume increase in the subsurface but involved fluids are also chemically aggressive to concrete constructions, while generating secondary ettringite formation in contact with cementitious materials. This influences the choice of concrete mix design for building components embedded in black shale.

Today's routine geotechnical studies addressing the black shale hazard include a series of expensive time-consuming quantitative analytical modal composition studies involving e.g. X-ray diffraction (XRD), scanning electron microscopy (SEM), and geochemical profiling of the sulfur content. However, Heintz et al. (2013)

postulated already that the use of in-situ tools like the MENARD pressuremeter to evaluate swelling risk of black shales indirectly is time-saving, pragmatic and equally suitable for this geotechnical risk assessment: pressuremeter limit pressure (pl_M) values above 1.2 MPa have been statistically linked to a high pyrite-alteration-induced swelling potential, while values below 0.7 MPa suggest minimal risk.
Following this first approach presented by Heintz et al (2013), we discuss in this paper the updated laboratory and pressuremeter results.

2. Geological / Geotechnical Background

2.1. Toarcian Black Shales and Pyrite Content

Posidonia shales, are mapped over a large part of southern Luxembourg between Pétange in the west and Bettembourg in the east are outcrops of the north-eastern periphery of the Paris Basin.
Posidonia shales are also known in Luxembourg as “Schistes-bitumineux” and/or “schiste-carton” and refers to the lower part of the marly layers of the Upper Lias (Lower Jurassic) and is stratigraphically subdivided into the units “Couches à Harpoceras falciferum” (lo1) and “Couches à Hildoceras bifrons” (lo2). They consist mainly of dark grey to black, subhorizontal laminated, mostly bituminous clay-rich marlstones in unweathered conditions. The total thickness of this Toarcian unit is about 50 meters in Luxembourg. Sedimentologically, it is interpreted as an alternating strata of mainly bituminous, fossil-rich, thin mudstone and marl layers to banks, which depending in the degree of weathering contain on average < 20% lime and with lesser finely laminated hard limestones. Pyrite occurs finely distributed and/or in nodules within the unit. As the stratigraphic names indicate, the rock is very rich in fossils and is actually world-famous for its paleontological records.
At the time of deposition of these formations, the sea was deeper compared to the underlying middle liassic sandstone (lm3; Grès médiolassique), who had been characterized by continental sand deposits. The Posidonia shale deposits is significantly deeper marine and sedimenting organisms were gradually buried by layers of clay rich mud, so that the organic matter was slowly transformed, in the absence of oxygen, into kerogen. Based on the reducing conditions the iron present in porefluides could only crystallize as pyrite, during diagenesis while reacting with sulfur released during the anaerobic transformation of the organic matter. Sulfide minerals are therefore petrographically often observed near fossils.
Presently a quaternary Posidonian shale alteration layer about 11-13 meters thickness is recorded in the study area, characterized by pressuremeter limit pressure pl_M < 5 MPa and deformation moduli E_M < 100 MPa. Close to the surface, at a depth about 3 meters, the altered black shales appear as light brown to yellowish plastic soils. Only below this level the grey tones and he fine layering gradually becomes visible in the drill cores with depth.

2.2 Swelling Mechanism

We will report here under sub-point (a) the mineralogical geochemical parameters of the swelling mechanism and under (b) the geotechnical constrains.
a) The exact pyrite alteration that leads to the swelling of the black shales is, as already mentioned, very complex (cf. Rimstidt & Vaughan, 2003) and can have different catalysts involved. Furthermore, it is still debated whether the resulting oxygen in the sulfate and oxyhydroxide product is derived from water and/or dissolved oxygen (see Eq.: 1 and 2). Thus, the exact intermediate bio-geochemical reactions and/or their importance are still a matter of scientific debate (e.g. microbiological involvement of Thiobacillus ferrooxidans and Thiobacillus sp.). Following from this, an exact petrological description of the processes would clearly go beyond the scope of this geotechnical paper. Thus, we focus here on the widely accepted fundamental geochemical reactions during the alteration of pyrite-rich black shales.
Pyrit alteration to goethite and new formation of gypsum in close association to goethite has been microscopically documented on Luxembourgian black shales by Cappuyns et al. (2019). They further defined, that the acidity generated due to the sulphuric acid produced by the pyrite oxidation is directly neutralized with calcite dissolution. And resulting crystallizing gypsum has been observed to partly replace oxidized stratiform pyrite deposits. Based on these observations it is possible to geochemicaly describe the Toarcian black shale pyrite alteration reaction as follows:

(1) $FeS_2 + 7\,O_2 + 2H_2O \rightarrow 2FeSO_4 + 2H_2SO_4$
(2) $4FeSO_4 + O_2 + 2H_2SO_4 \rightarrow 2Fe_2(SO_4)_3 + 2H_2O$
(3) $Fe_2(SO_4)_3 + 6H_2O \rightarrow 2Fe(OH)_3 + 3H_2SO_4$
(4) $CaCO_3 + H_2SO_4 + H^+ \rightarrow CaSO_4{\cdot}2H_2O + CO_2$

These chemical reaction equations which are starting with pyrite (FeS_2) and after oxidation are resulting in gypsum $CaSO_4{\cdot}2H_2O$ reflect a complex interaction of solids with water and oxygen generating volume increase and swell pressures like mentioned before. A simple test to substantiate the oxidation theory consists of putting shale samples in glass jars which are sealed. It is reported that subsequent analysis of the air in the jars showed the oxygen content to have dropped from a normal 20.7% to as low as 12% (Engineering News-Record, 1960).
b) From a geotechnical point of view, the alteration process can be characterized as follows: the more advanced the weathering process is, the more intensive the oxidation of black shale has been, the more complete the conversion of pyrite into gypsum has been, and the lower the pyrite content will be, which are finally resulting in a lower risk of swelling and damage to buildings.

3. Assement of the swelling risk using pressuremeter test

The decreasing degrees of weathering with depth, characterized by soft black shales soils near the surface, evoluating to unweathered compact black shale rocks, are determined by increasing limit pressures pl_M from pressuremeter tests. The pyrite content at the test depths can be determined on the corresponding drill cores by laboratory tests. By correlating the pressuremeter limit pressure pl_M and the corresponding pyrite content, the swelling risk can be estimated through cored pressuremeter boreholes (Heintz et al. 2005) as shown in the histogram presented in Fig. 3 and the associated table of values.

The qualitative denomination for consolidation in French, according to the Fascicule 62 (modified) and NF P94-261 classification systems, have not been translated to avoid confusion with foreign classification systems.

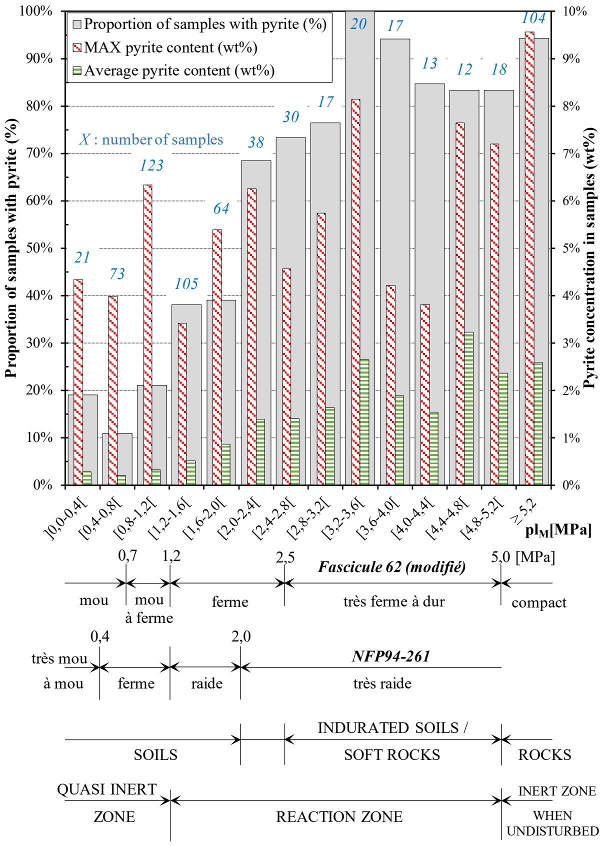

Fig. 3. Evolution in pyrite content as a function of the pressuremeter limit pressure pl_M (Data in Table 1)

Table 1: Sample set used for Fig. 3 reporting the number of samples for each pl_M group and their pyrite content.

pl_M (Mpa)	Number of samples (X)	Samples with pyrite	Proportion of X with pyrite (%)	Pyrite concentration		
				min (wt%)	MAX (wt%)	average (wt%)
0.0 - 0.4	21	4	19	0	4,34	0,28
0.4 - 0.8	73	8	11	0	3,98	0,2
0.8 - 1.2	123	26	21	0	6,34	0,33
1.2 - 1.6	105	40	38	0	3,42	0,52
1.6 - 2.0	64	25	39	0	5,39	0,87
2.0 - 2.4	38	26	68	0	6,26	1,39
2.4 - 2.8	30	22	73	0	4,57	1,41
2.8 - 3.2	17	13	76	0	5,74	1,64
3.2 - 3.6	20	20	100	0,24	8,15	2,65
3.6 - 4.0	17	16	94	0	4,21	1,89
4.0 - 4.4	13	11	85	0	3,81	1,54
4.4 - 4.8	12	10	83	0	7,65	3,23
4.8 - 5.2	18	15	83	0	7,2	2,36
≥5.2	104	98	94	0	9,56	2,6

The used sample set includes 655 pressuremeter tests in combination with XRD modal laboratory measurements of Posidonia shales on the territory of Luxembourg performed during the last three decades.

A first order observation is, that the increasing pl_M values in the histogram can be statistically correlated down hole with increasing depths.

The observed average pyrite concentrations (horizontally striped bars in Fig. 3) can be discriminated into three different categories of pyritic swelling risk:

a) a near-surface, *quasi-inert zone* of highly weathered soils, characterised by $pl_M < 1,2$ MPa, in which the average pyrite content has been reduced by oxygen attack to a minimum average content of < 0,33wt% (% in weight). The numbers above the bars in fig. 3 show an increased number of samples taken around the threshold value of $pl_M = 1,2$ MPa; the results confirm this value as a threshold which is a major target of the present study. In the quasi-inert zone swelling is very rare and occurs only in exceptional conditions.

b) A *reaction zone* starting at $pl_M = 1,2$ MPa, with a down hole increasing tendency of fresh pyrite content with depth. This depth profile is due to the top-down weathering front penetrating into the rock formation with time and resulting in the systematic, still ongoing pyrite to gypsum transformation.

If black shale soils producing $pl_M > 1.2$ MPa are incised by construction works, geotechnical measures are essential to prevent resulting uncontrolled swelling processes. These measures to prevent uplift damage are considered to be essential for construction projects. Especially for sub-surface constructions and buildings with several basement levels.

At the base of the reaction zone, the average pyrite concentration reaches a maximum of 3,23 wt% in indurated black shale soils producing a $pl_M = 4{,}7$MPa, i.e. close to $pl_M = 5$ MPa, the conventionally defined limit at the transition from soil to rock.

c) The rocky black shale, which generates $pl_M > 5$MPa is considered *inert in undisturbed conditions*. Presently the alteration reactions have not started within the rock but can at any moment start if the needed physico-chemical parameters are installed. So this rocky black shale type has the highest - still initial - swelling potential.

4. Damage events in the risk zones below and above $pl_M = 1.2$ MPa

The presence of pyrite, even in heavily weathered black shales of the quasi-inert zone, can never be ruled out (see Table 1), but the risk of uplift damage for constructions embedded in this zone is minor to negligible as the pyrite content is widely distributed in lowest concentrations: according to Fig. 3 only 11 to 21% of the samples (grey shaded bars in Fig. 3) examined from soils producing $pl_M < 1,2$ MPa contained small amounts of pyrite. It can therefore be expected that positive volume changes due to the pyrite alteration is amortised by the plastic behaviour of these weakly consolidated soils.

This further explains why heaving construction damage of single-family houses, which would be attributable to pyrite swelling in the quasi-inert zone is very rarely known in Luxembourg. In general, these houses are adapted to the natural morphology and there foundation

depth usually lies above the base of the quasi-inert zone, whose limit value pl_M = 1.2 MPa can be statistically correlated with a depth about > 4 meters.
Consequently, it is a matter of the geotechnical engineering judgment whether precautionary measures against oxygen attack of the black shale are appropriate for projects embedded in the quasi-inert zone.
With reference to Penner et al. (1970) however, who reported heat-induced heave damage in soils with only 0,1wt% pyrite content, the introduction of heat into the pyrite-containing subsoil should be prevented.
However, when cutting into the reaction zone, the risk of swelling increases significantly with depth, especially in the layers producing pl_M > 3.2 MPa (Fig. 3) in which the pyrite is distributed very narrowly and in high concentrations: up to 100% of the samples can contain an average of over 2.5 per-cent pyrite by weight.
Also in rocky black shales whose pl_M, produced by high pressure pressuremeter tests (Arsonnet, 2014) is up to 15 MPa, the interlayer growth of gypsum crystals in the fine stratification plains, begins shortly after its exposure due to oxidation of the pyrite. This leads to cracking and uplift of the fine layered structure, which affects the foundations and causes considerable construction damage.

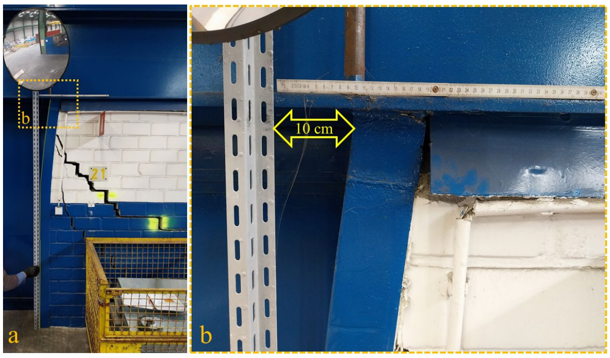

Fig. 4: a) Photo illustrating the deformation of an IPE200 steel beam due to gypsum crystallisation as a result of pyrite oxidation of the subsoil consisting of rocky black shales, characterised by pl_M>5 MPa; b) Zoompicture of the deformed IPE200 steel beam. (Photos: Eurasol, 2025)

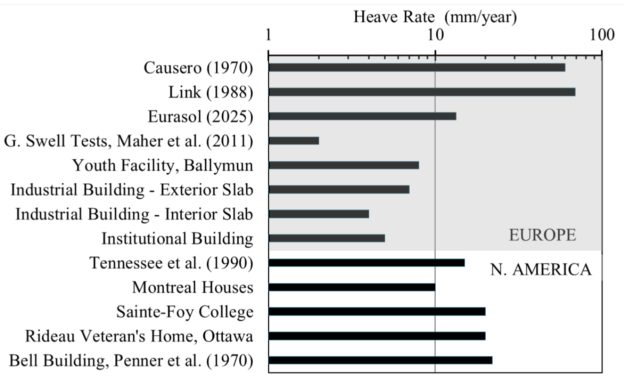

Fig. 5: Recorded heave rates in buildings in Europe and N America due to pyrite induced heave (modified from Maher and Gray, 2014 with references therein and updated with new data).

An extreme example (Fig. 4) shows the deformation of an IPE200 steel beam in a hall in an industrial area that was levelled over a large area of black shale.
As a result of the ongoing uplift movements, up to 13,3 mm/year comparable to the measurements summarised by Maher and Gray (2014) in Fig. 5, the hall was declared unfit for use. Planning for the rehabilitation of the hall is currently underway (Eurasol, 2025).

5. CONCLUSIONS

Even if the swelling hazard is considered to be negligible, the foundation ground in pyritic black shale should always be protected against oxygen attack as a precautionary measure. A barrier layer of overlapping plastic sheeting is often sufficient, the integrity of which must be maintained during the construction phase. The financial expense of this preventive measure is negligible in relation to the corresponding cost of rehabilitation measures in the event of heave damage.

Table 2: Red-yellow-green classification of specific construction requirements based on pl_M and pyrite content for foundations in black shale formations.

Black shale type	pl_M (Mpa)	Pyrite content	Swelling hazard	Spec. construction
1. Quasi-inert zone	<1.2	+/-	+/-	+/-
2. Reaction zone	>1.2 - ≤ 5	+ / ++	+ / ++	suggested / needed
3. Rocky black shale zone	>5	+++	+++	needed

A greater protection against heave damage is provided for example by the design of a crawl space in which the black shale can expand freely. However, if the spans of the unsupported slab above the crawl space become too large, the consideration of the heave problem shifts to the dimensioning of piles.
The depth of the reaction zone within which the heave friction acts on the pile shaft during the service life of the construction depends primarily on the situational oxygen availability to be estimated and the not clearly defined portion of swelling that can be responsible for pile lift. The note by Chen (2000), which states that for expansive clays 15% of the swelling pressure can be taken as uplift along the pile shaft, can serve as a guide both here and for anchoring. The balancing friction capacity below the reaction zone can then be calculated as usual using the friction values, which can be determined from the relevant Eurocode 7 tables or other pertinent standards as a function of pl_M.
Furthermore, the introduction of heat into the pyritic black shale should be avoided as far as possible by means of adequate insulation. Heat acts as a catalyst in the chemical reactions that lead to uplift.
The third main precaution is to prevent oxygen attack on the black shale foundation ground by drainage, which requires the application of appropriate sealing measures for buildings, taking into account the build-up of water pressure in the black shale excavations which can be regarded as impermeable.
Construction aggregates suspected of containing pyrite, especially if they consist of excavated black shale, should

be analysed using appropriate laboratory tests, before introducing their way into built environment (Maher and Gray, 2014).

For foundations exerting pressures > 0.3 MPa (Tietze 1981) and approximately 0.4 MPa (Causero 1970) respectively, no uplift damage should occur, according to the observations of these authors; these values can therefore be regarded as indicative, at least for the geotechnical design related to black shales outcropping in the Paris Basin and the South German Basin. However, caution is advised with regard to Fig. 2.

In geotechnical engineering threshold values should generally not be seen as a black box discrimination tool excluding thorough geological and geotechnical investigations of the local geology and damage report research.

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