PYRITIC HEAVE IN IRELAND: THE ROLE OF PETROGRAPHY

Fred H. Shrimer,
Golder Associates, Vancouver Canada

Alan V. Bromley,
Cornwall, UK

ABSTRACT

Hundreds to thousands of cases of heave of structures have been attributed to the presence of pyrite-bearing aggregate in Ireland in recent years. Leading to numerous lawsuits, expenses in the millions of Euros, and untold emotional toll on thousands of people and their families, this problem was defined through the use of petrography of the aggregates at the centre of the problem.

This paper examines the use of petrography in the investigations that were undertaken, and discusses the means that were used to assess the nature of the problem, as well as the differences found in the Irish cases of heave compared with those in other countries. In addition, due to the fine-grained nature of the aggregates, we look at the complementary analytical techniques that were used to augment the petrography.

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<u>INTRODUCTION</u>

Beginning in 2006, damage was being noticed in recently-constructed buildings, mostly residences, in north Dublin, Ireland. The damage being reported consisted of cracking in interior walls, distortion of door and window frames, cracking of windows, rising and unevenness of floors, cracking of concrete floors, and cracking of exterior finishes. The affected buildings were generally about 2 to 4 years old at the time that these defects were observed and reported.

Structural engineers were initially brought in to assess the damage and to provide remedial solutions. At first, the engineers' perspective was that the differential movements might be explained by settlement of structural elements, thus providing a means of upward movement of the floor slabs relative to wall, the floor slabs having been constructed on fill aggregates placed on native soil subgrade. However, after the cracks had been repaired and doors trimmed to open and close smoothly, the same damage recurred in the repaired areas within about six months.

Photograph 1: Typical damage observed in houses: cracks and bowed-out walls (left) and cracked heaved floor (right)

Since the structural engineers were unable to provide suitable explanations of the nature of the damaging mechanisms that were affecting the structures, geotechnical and geological experts were consulted to assess foundation conditions and the degree of compaction of supporting fill beneath the concrete floor slabs. The geotechnical engineers were able to confirm that settlement of the structures (due to poor soil conditions, for example) was not at issue.

The focus of the investigations then turned toward the nature of the fill aggregates that supported the slabs-on-grade. Geologists examined samples of the infill aggregate, and determined that the likely mechanism which had created the deterioration observed within the buildings was "heave", which is an upward movement of structural components due to expansion of the structural fill aggregates.

In terms of the effects of pyrite-induced heave, the original investigations–conducted between 2006 and 2008 -- indicated that several hundred homes in the north Dublin area were affected by the problem. Subsequent to the earlier work, estimates are that up to 20,000 buildings, primarily residences, could be affected.

Hundreds of claims were made by homeowners against their home insurance policies and numerous lawsuits were launched against both builders/developers as well as quarry operators/owners, to obtain compensation for the damages. Due to the progressive, cumulative and irreversible nature of pyritic heave, the only failsafe solution has consisted of the removal of the concrete floors, the excavation and disposal of the pyrite-bearing aggregate, the replacement with suitable fill, and the reinstatement of the slabs, along with cosmetic restoration of the properties. On average, the costs of undertaking this work ranged from €40,000 to 90,000 per residence/structure.

GEOLOGY OF THE AGGREGATE

The aggregates that were utilized as infill were mainly sourced from a quarry located on the north side of Dublin. The quarry began production in 2003. It was estimated that some

Photograph 2: View of quarry face, showing thinly bedded mudstone strata

2,000,000 tonnes of aggregate were produced and shipped from this quarry prior to 2008; at the time of writing, only about 5% of the total production has been accounted for. According to Jones, Somerville and Strogen (1988), the rocks belong to the Tober Colleen formation (Lower

Carboniferous), which generally includes limestone, siltstone, mudstone, shale and sandstone. The rocks are typically thin-bedded and interlayering is common.

The Carboniferous rocks in the Dublin area have a southerly dip: the older rocks are exposed in the north. An older working, about 1 km south of the quarry of interest, produced high-quality limestone aggregates. In the quarry of interest, the aggregates produced are composed of about 85% calcareous mudstone and siltstone, and only 15% of limestone, sandstone and sandy limestone.

MANIFESTATION OF PROBLEMS AND INITIAL INVESTIGATION

Within a few years of construction, residents of several large estates constructed starting in 2003 noticed that cracks had developed in their walls (mainly around door and window openings), that floors seemed uneven, and that doors were sticking. After structural engineers had attempted to diagnose and address the issues and their causes, and ruled out issues related to workmanship and concrete shrinkage, geotechnical engineers and geologists were consulted to assess the performance of the foundation system and the slab-supporting fill aggregates.

After determining that geotechnical issues such as settlement or failure of foundation systems were not at fault, focus was turned towards assessment of the infill aggregate that supported the concrete floor slabs. Holes were drilled or cut through walls and floors to access and sample the fill; these samples were then subjected to engineering and geological assessments.

PETROGRAPHIC EXAMINATION

Samples of infill aggregate were subjected to Petrographic examination, following the methods given in ASTM C 295 and CSA A23.2-15A. We found that the material was composed dominantly of poor quality calcareous mudstone and siltstone, with minor amounts of limestone and calcareous sandstone. In general, the rock was comprised of a mixture of clay- and silt-sized grains, with a few sand-sized grains (mainly quartz); as a consequence, it was termed "Mudstone" to reflect rock formed of a mixture of fine-grained sediments of clastic origin. Microcrystalline calcite is dispersed throughout in variable amounts; hence "calcareous mudstone".

Calcareous mudstones and siltstones were generally well-laminated though some siltstones were more massive, lacking obvious lamination.

Mineral constituents include clays, fine-grained mica, chlorite, quartz and calcite with subordinate dolomite, iron oxides and lithic fragments. Pyrite is a ubiquitous minor component though it is commonly so fine-grained that it can only be recognised under the ore microscope or by scanning electron microscopy.

Secondary gypsum, aragonite and calcite were ubiquitous. They coated the surface of aggregate fragments and also grew along internal lamination planes.

Photograph 3: Mudstone aggregate particles, after having been broken to expose interiors that contain numerous secondary gypsum deposits

To assist in classifying the aggregate's engineering quality, we used the "Petrographic Number" (PN) method, commonly employed in Canada. A PN of 250 was determined for the first sample of fill aggregate examined in early 2007. This placed the sample in the range of "very poor" quality, relative to the scale most commonly used for PN classification of samples:

PN : 100–140 = Good
PN: 140–165 = Fair
PN: 165–200 = Poor
PN: >200 = Very poor/marginal/unsuitable

Subsequent samples of aggregate, also from the same quarry source, were tested from a variety of buildings; the samples had PNs that were generally in the 190–320 range.

Physical-mechanical testing was also carried out on the samples, as part of the larger assessment of the aggregate. Results are summarized below.

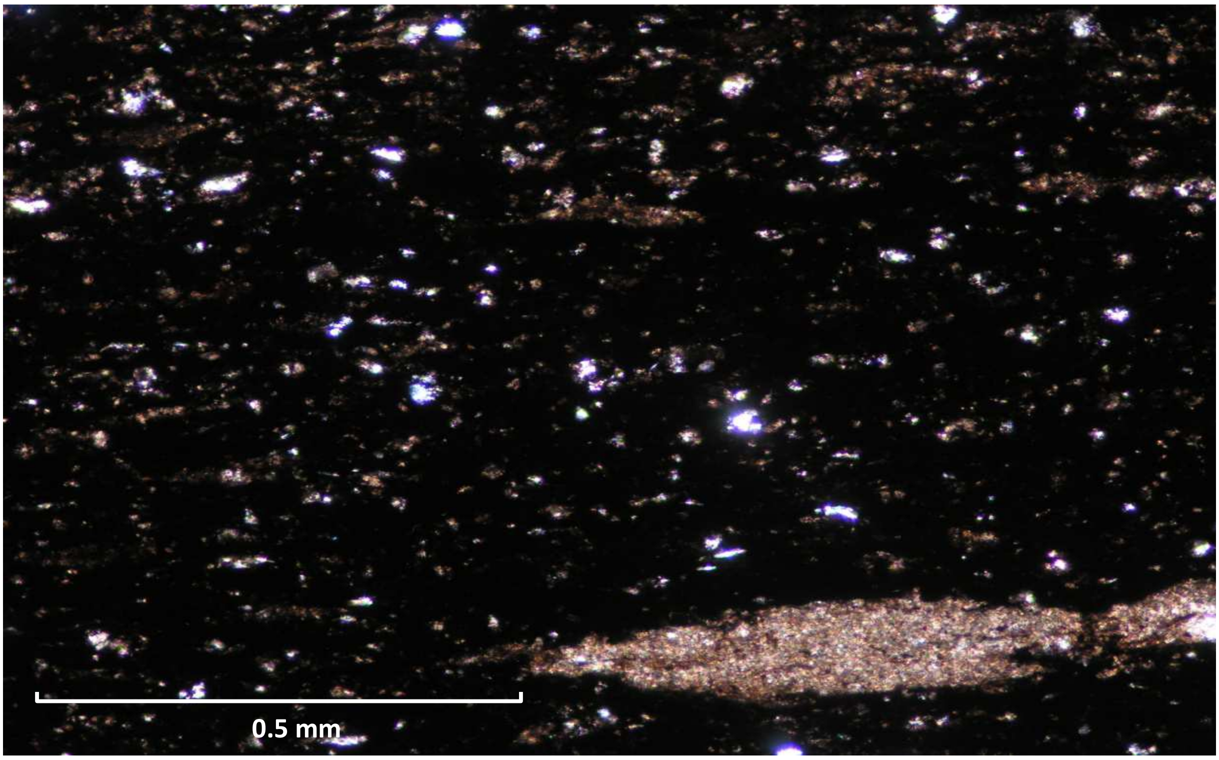
Table 1: Physical test results of Mudstone aggregate

Test	Property	Result	Rating
Specific Gravity	Density	2.45–2.53	Fair
Absorption	Water/air uptake	4.5–7.6%	Poor
Micro-Deval	Abrasion	50–59% loss	Very poor
Soundness	Durability	20–67% loss	Fair - very poor
Ten Percent fines	Degradation	25–90 kN	Substandard

These results generally indicated that the aggregate was of poor mechanical quality, and was prone to degradation, attrition and breakdown.

Thin-Section Examination of Mudstone

Several thin-section specimens of mudstone were prepared for examination in the polarizing microscope. Due to the fine-grained nature of the rocks, sections were prepared to thicknesses of about 20 µm.

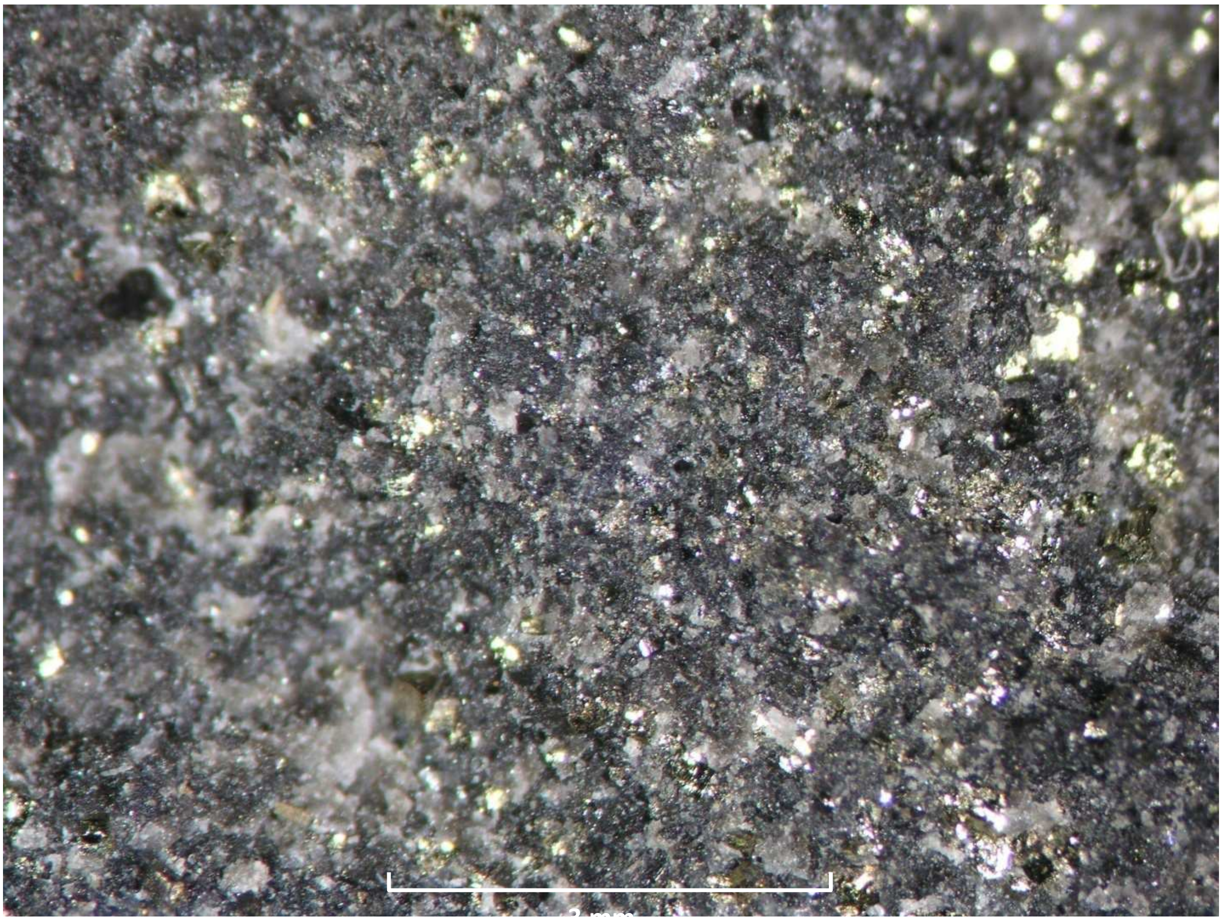


Photograph 4: Thin-section view in cross-polarized light: a dark and very fine-grained matrix, with detrital grains of quartz, calcite and feldspar

Many of the rocks were so fine-grained that conventional thin section petrography was of limited use. They all consisted principally of irresolvable clay minerals, fine-grained mica, chlorite and quartz in variable amounts. The lamination was defined by parallelism of clays and mica. They were cut by irregular veinlets of calcite. Fine-grained, disseminated pyrite was almost ubiquitous, albeit in minor amounts.

Pyrite

Some pyrite was visible with the unaided eye and under a low power stereomicroscope. It occurred as cubes, pyritohedra and irregular grains and stringers.

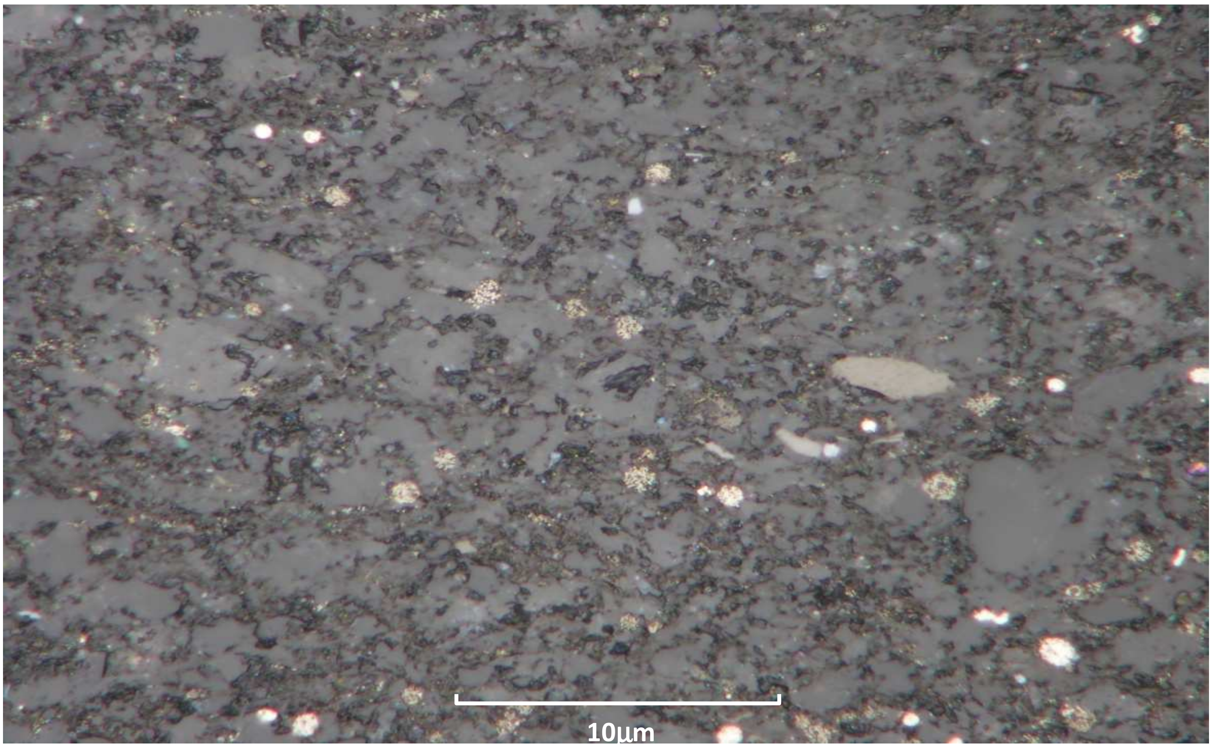


Photograph 5: View of mudstone particle interior showing crystalline pyrite in a fine-grained matrix of clay- and silt-sized grains. Scale bar = 6 mm.

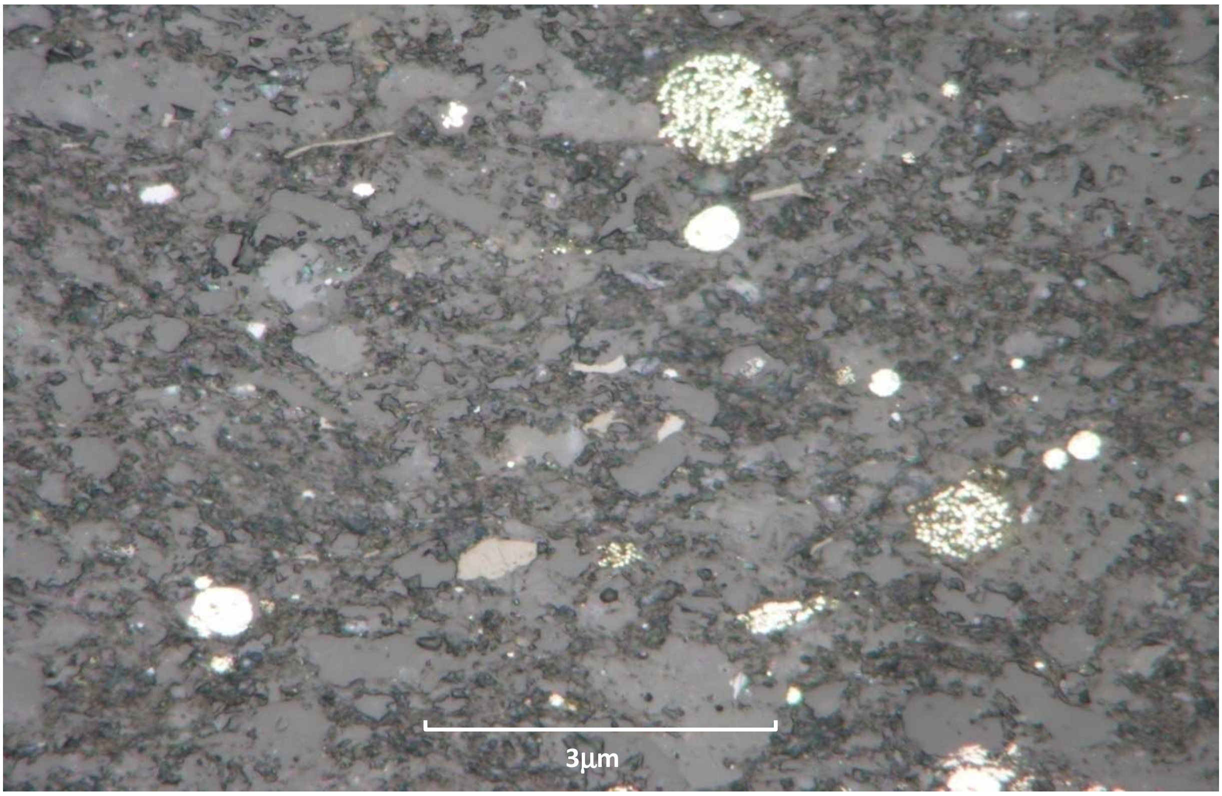
The morphology, size range and distribution of pyrite was investigated by reflected light polarising microscopy, augmented where appropriate by scanning electron microscopy (secondary electron and back-scattered electron images).

Pyrite crystals varied in size between a few millimetres and less than 1 micrometre. Crystals less than approximately 25 micrometres in size predominated. They varied in shape between idiomorphic (cubes and pyritohedra), spheroidal and granular. Framboids, made up from solid or hollow spheres of minute spherical grains, were common. Typically, they were < 50 micrometres in diameter.

Pyrite may be dispersed, more or less uniformly, in mudstone and siltstone fragments though sometimes it is concentrated in veins and stringers parallel with bedding planes.

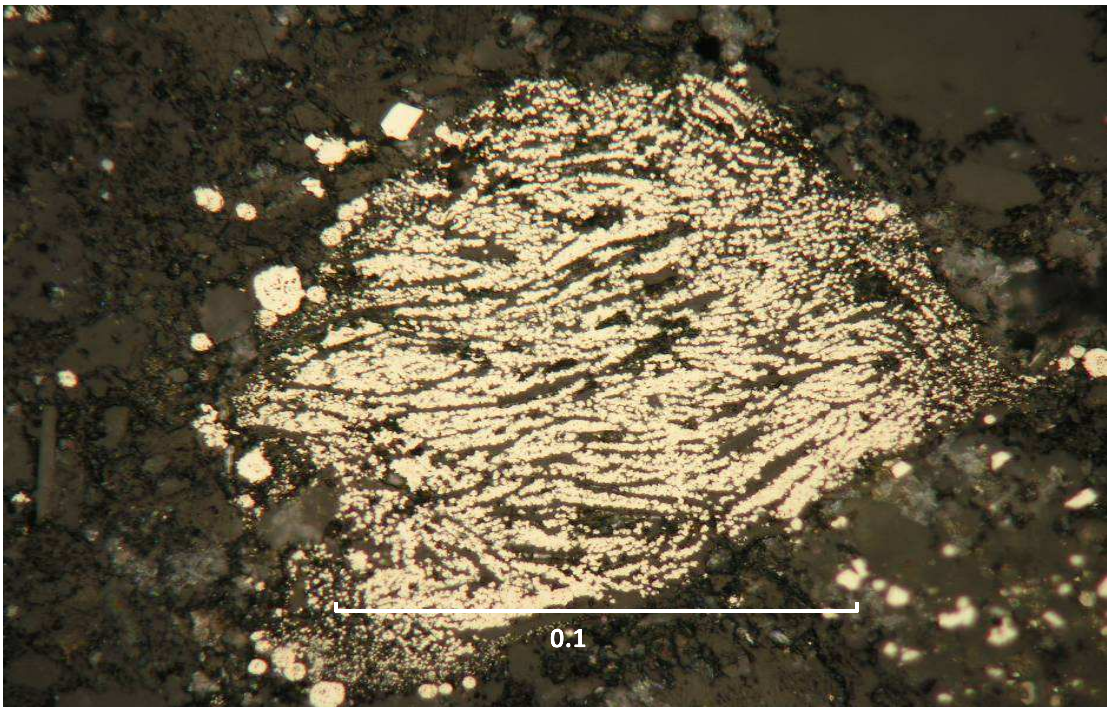


Photograph 6: View in reflected light of a thin-section of mudstone. Clusters of framboidal pyrite are observed.



Photograph 7: Framboids of pyrite in mudstone

Pyrite content, determined chemically and by modal analysis, varied between *circa* 5% and less than 0.5%. Average original pyrite content of the aggregate as a whole was between 1% and 3%.



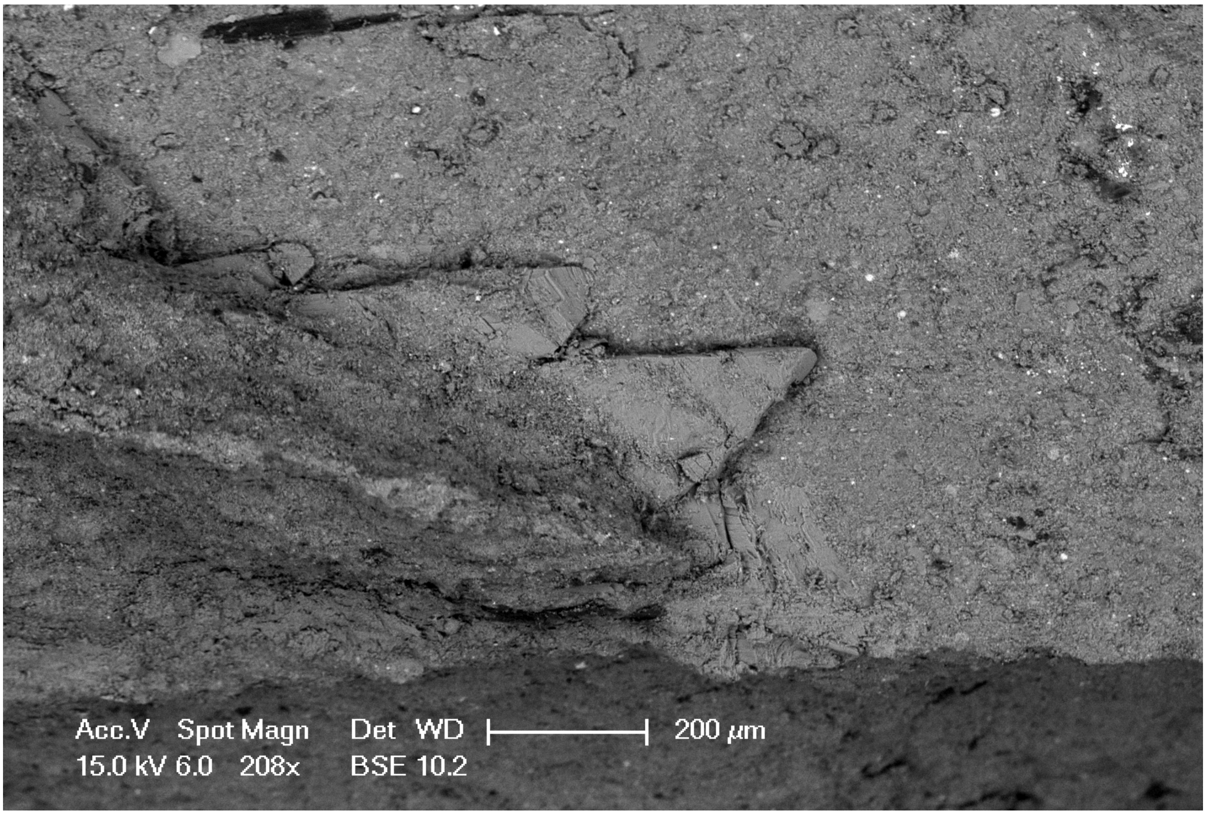
Photograph 8: View of fine-grained and framboidal pyrite in mudstone

Between *circa* 1% and 10% of pyrite was converted to brown iron oxide (limonite *sensu lato*). Usually, whole crystals are pseudomorphed and sometimes enclosed by haloes of dispersed oxide. Fresh pyrite and oxidised pyrite were commonly juxtaposed in the same fragment.

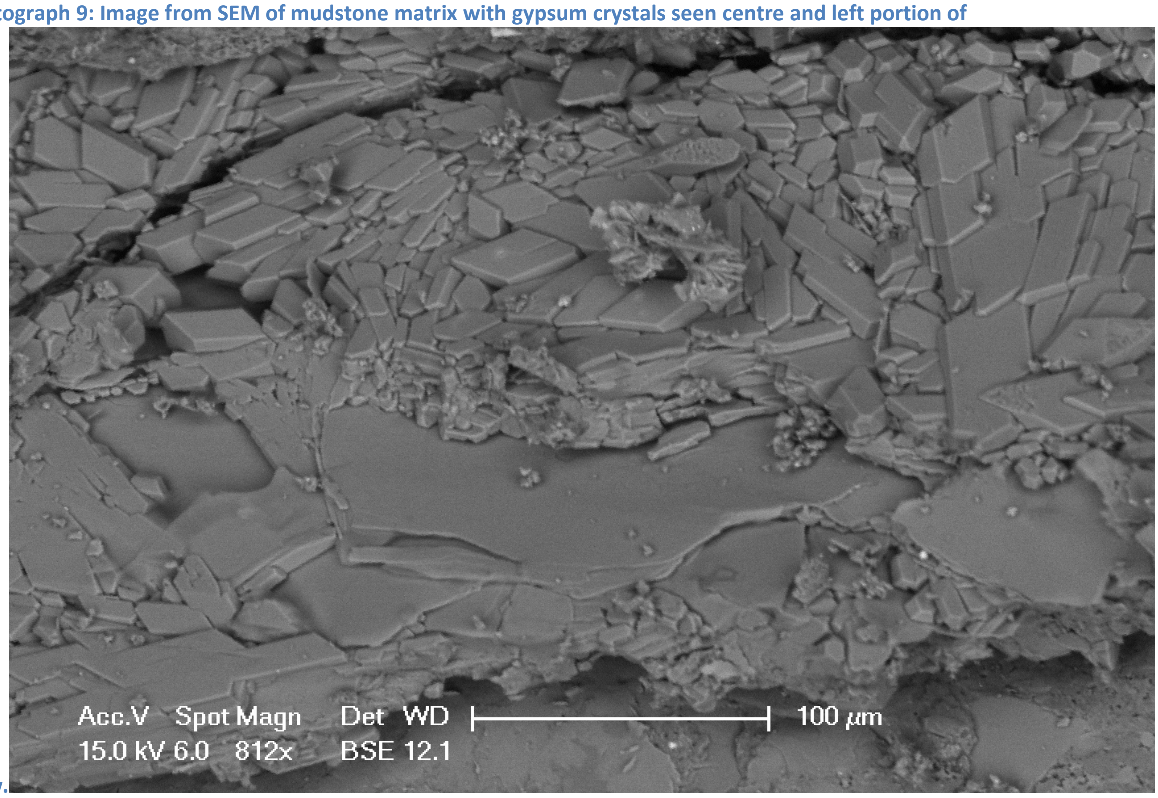
SCANNING ELECTRON MICROSCOPY

Untreated hand specimens and polished thin sections of aggregate were investigated by scanning electron microscopy (secondary electron imaging and back-scattered electron imaging). Mineral chemistry and element distribution mapping supplemented the energy dispersive X-ray analysis. Rietveld analysis was used to supplement conventional modal analysis in the estimation of mineralogical composition.

Scanning electron microscopy of untreated specimens offered considerable advantages over optical microscopy in the investigation of delicate surface coatings on aggregate fragments and in the study of soluble minerals, principally gypsum, which were modified or in some cases dissolved away during the preparation of petrographic thin sections.



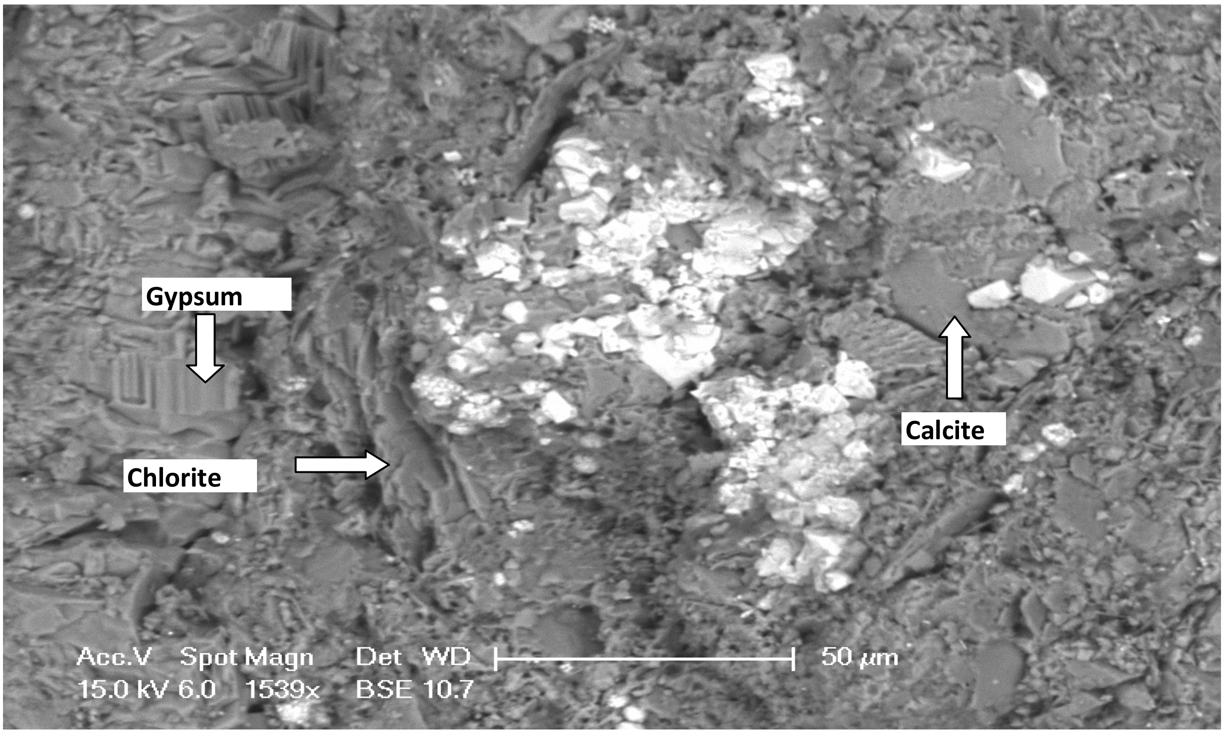
Photograph 9: Image from SEM of mudstone matrix with gypsum crystals seen centre and left portion of view.



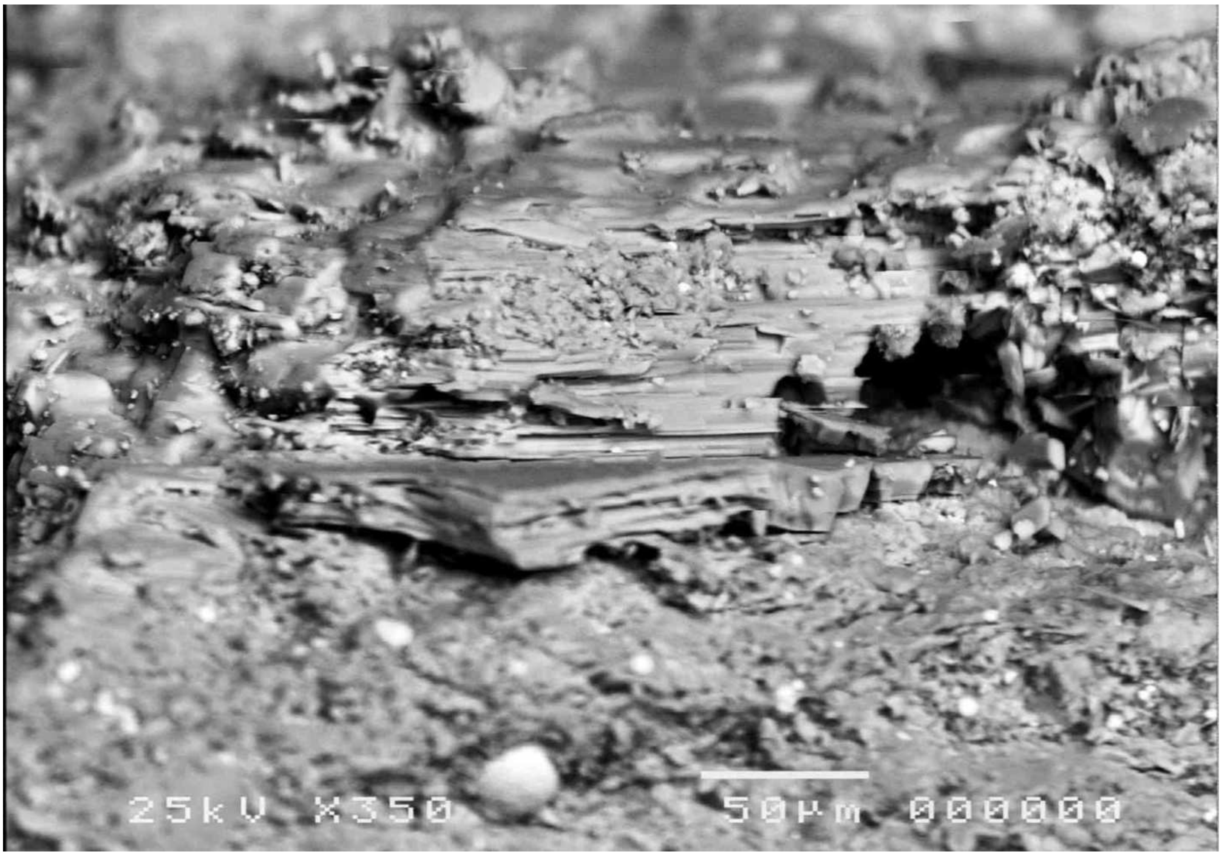
Photograph 10: Gypsum crystals seen on mudstone interior surface

In the SEM, we were able to determine with greater precision the composition of the rocks, their texture and structure, the nature of the secondary minerals, and other features of relevance in the investigations.

Photograph 11: Groups of gypsum crystals seen on a lamination surface of a mudstone particle.



Photograph 12: Surface of a particle interior, showing pyrite (light coloured crystals) in the central portion of the image, adjacent to a vein of chlorite to the left, gypsum further to the left, and calcite to the right.



Photograph 13: Gypsum crystals and pyrite framboids in mudstone.

SEM investigation revealed that secondary gypsum and aragonite were far more abundant than initially suspected, following optical microscopy alone.

Gypsum had two modes of occurrence:

1 It formed partial crusts of idiomorphic crystals and clusters on the surface of aggregate fragments where it was commonly overgrown by delicate, acicular crystals of aragonite ($CaCO_3$).

2 It grows as layers of fibrous crystals or rosettes of idiomorphic crystals along bedding planes. Clearly, growth of secondary gypsum has caused spawling and expansion of individual aggregate fragments normal to bedding. Bulk linear expansion of up to 0.2% was measured in several large aggregate fragments.

SUPPLEMENTARY ANALYSES

Energy-Dispersive X-Ray microanalysis

Energy-dispersive X-ray microanalysis (EDMA) was used to confirm the identification of sulphides and secondary products formed during oxidation.

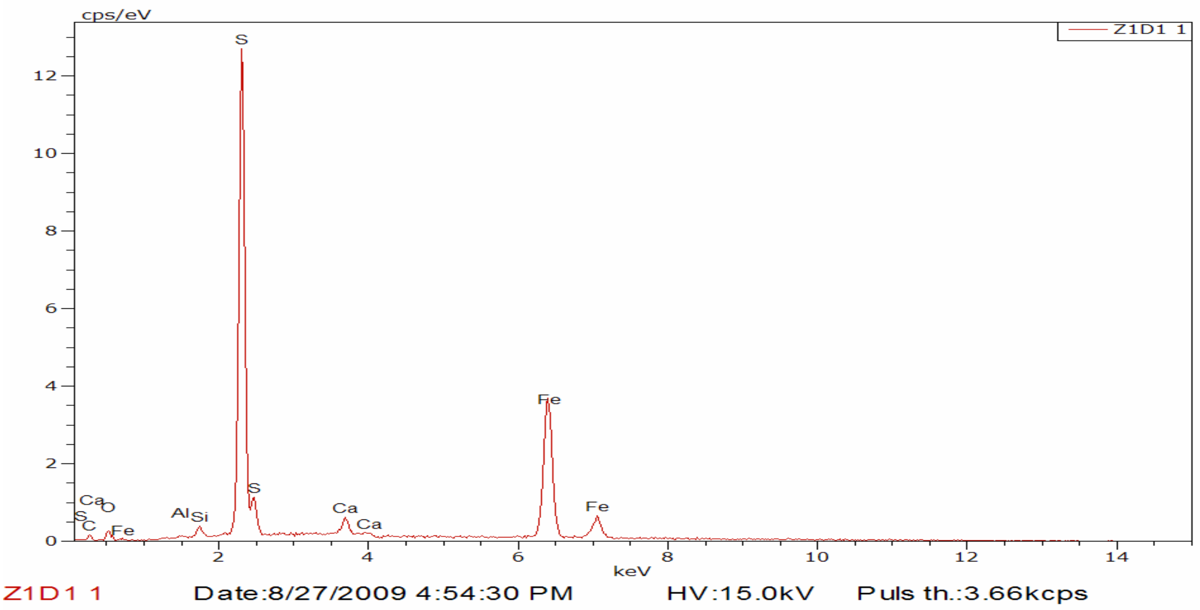


Figure 1 EDX scan of a crystal observed in the SEM examination, illustrating strong peaks for Sulphur and Iron, consistent with pyrite's composition

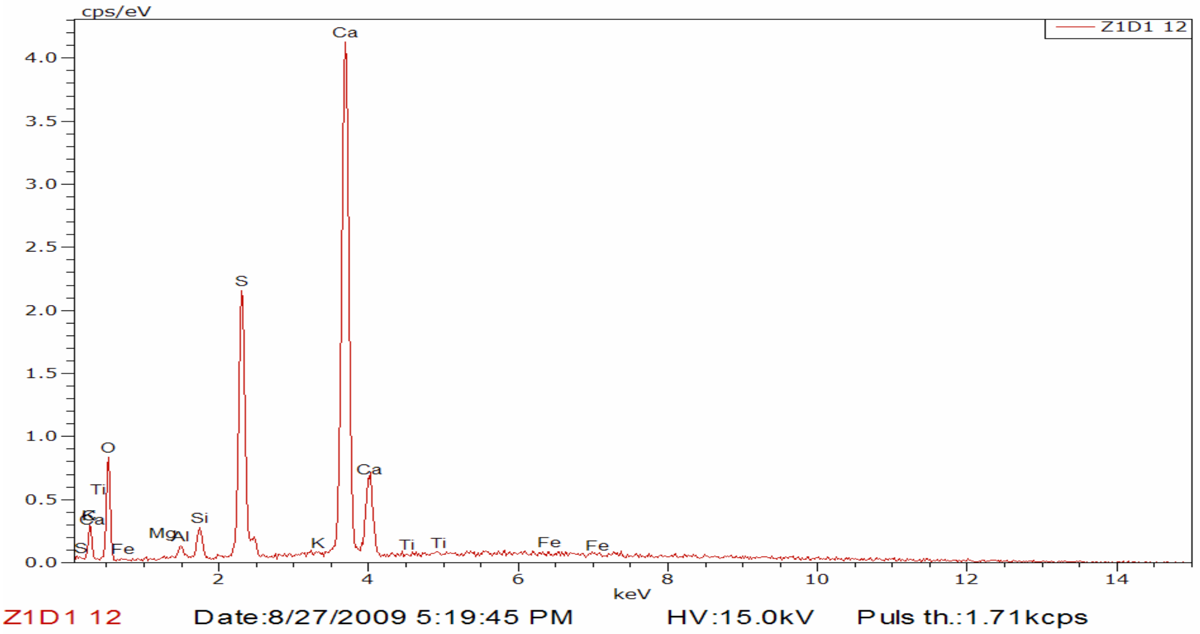


Figure 2: Strong peaks for Calcium, Sulphur and Oxygen indicative of Gypsum

Element Mapping

Digital element mapping was used to illustrate the distribution of various phases in the altered aggregate. Figure 16 clearly shows a vein of gypsum (high Ca + S) in calcareous mudstone with principal components (Ca, Si, Al and Fe) contained in calcite, quartz, clay minerals and chlorite.

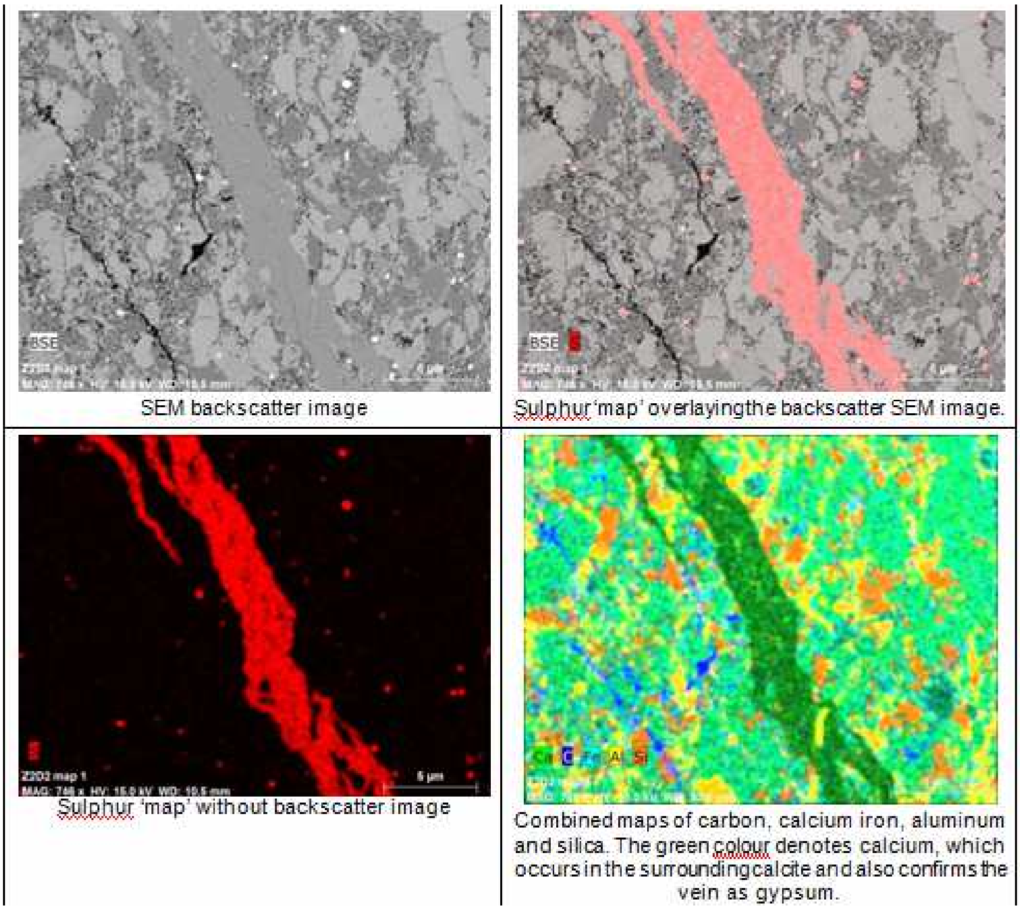


Figure 3: Imagery from element mapping of a portion of a mudstone particle, confirming the presence of gypsum in a vein.

MECHANISM OF DAMAGE

The petrographic investigations strongly indicated that expansion of the aggregate and heave of newly-laid floor slabs in the north Dublin area is a result of *in situ* oxidation of fine-grained, commonly framboidal pyrite in laminated calcareous mudstones, which led to the production of expansive gypsum within and throughout the aggregate mass.

Pyrite is stable under reducing conditions but when exposed to moist air it undergoes oxidation with the formation of soluble sulphur species (essentially sulphuric acid). Fine-grained and especially framboidal pyrite with high surface area/volume ratio is particularly susceptible to rapid oxidation. Even in the dry atmosphere of laboratories and museums, pyrite may decompose completely in just a few weeks.

The conversion of pyrite to iron oxide is expansive, but in these rocks much more expansion occurred when sulphuric acid reacted with calcite in the rock to form gypsum.

The total expansion of the mass of aggregate is a combination of two factors.

1 Inter-particle growth of gypsum causing aggregate fragments to be forced apart, and

2 Intra-particle growth of gypsum causing bed-normal expansion of individual aggregate fragments.

Photograph 14: Abundance of gypsum crystals on an opened lamination plane within a mudstone particle. Scale bar = 4 mm

The aggregates that supported the concrete slabs were placed to a thickness that was usually on the order of 0.5 - 1.4 m, and were typically sized to be about 75 mm (3") maximum nominal size; they were compacted prior to construction of the concrete slab.

The forces exerted by the growth of gypsum within the mass of aggregate thus were sufficient to lift ('heave') the slab, which, being unreinforced (as is typical of residential construction in Ireland), cracked. As differential movements thus occurred within the slabs, the walls, door frames, and window casements also distorted, and doors jammed; in addition, residents have reported windows cracking, water leakage and bent and breaking pipes within the walls. Confirmation of this mechanism is discussed in Maher *et al* (2011) where a large-scale laboratory swelling test was undertaken to assess expansive reaction of the Irish mudstone.

CONCLUSIONS

The Petrographic examinations were of central importance in diagnosing the cause of the problems and understanding how the heave was occurring (or had occurred), what was likely to occur in the future, and ways in which the problem could be dealt with. For example, the progressive and cumulative nature of the mechanism that enabled the mudstone aggregate to produce heave was something that was considered unlikely to slow down or stop for some

Photograph 15: Fragmented mudstone aggregate particle, illustrating the abundance of secondary gypsum crystal growth within the laminations of the rock.

considerable period of time, and thus, it was thought that the only sure means to deal with the problem was to remove the fill and replace it.

Additionally, comparison of the petrography of various samples of fill taken from hundreds of structures over a three-to-five year period enabled the determination that, in some cases, contractors had utilized aggregates from different source quarries–some crushed stone materials of good and sound quality had been mixed with poor quality, heave-prone aggregates, and thus the entire mass of fill was subject to removal and replacement.

Petrography provided the proof positive that the aggregates were of substandard quality in the sense that they were demonstrably not chemically inert, and that gypsum and other secondary minerals had developed both within the aggregate particles as well as on their surfaces. Petrography also determined that the nature of the deterioration was dependent upon the presence of (1) a weak mudstone rock of low durability (2) unstable framboidal pyrite in amounts greater than 1% and (3) calcite of sufficient proportion to enable the development of gypsum.

Until Petrographic studies had been completed, the nature of this serious and very expensive geological engineering phenomenon was not understood, and steps to deal with it–both in terms of repair as well as in revision of engineering specifications to address the issue–could not be taken.

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SUPPLEMENTAL PHOTOGRAPHS

Photograph 16: Bulging out distortion of walls, and cracking at door frames.

Photograph 17: View of a portion of north quarry face, showing laminated structure of mudstone.

Photograph 18: View of blast rock accumulation at foot of quarry face.

Photograph 20: Sample of mudstone aggregate as-received from subfloor investigation site. FOV about 30 cm.

Photograph 19: Mudstone particle split open on a lamination, exhibiting secondary gypsum crystal growth. FOV about 150 mm