

A laboratory study of the expansion of an Irish pyritic mudstone/siltstone fill material

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ABSTRACT

Extensive damage to dwellings in the east of Ireland, arising from the expansive pyritic fill material used beneath the floor slabs, prompted the laboratory study presented in this paper to quantify the expansion and study its influencing factors. A novel experimental arrangement, in which sample height and water supply were varied, was used to test a large quantity of active pyritic infill taken from an affected house in the greater Dublin area. The expansion of ten separate fill specimens was measured over a 175 day period from December 2010 to May 2011. Remarkably consistent results were obtained when the expansions were normalised by the sample height, with expansion rates of approximately 2–3 mm/year/m height of fill for specimens standing in water, where the rates were largely insensitive to the amount of water supplied. More rapid expansion was noted for one specimen to which no additional water was supplied during the test. A steady reduction in pH of the water, although relatively small, was consistent with pyrite oxidation reactions occurring in the samples during the test, and also implies the occurrence of leaching of ions from the pores in the mudstone and also that oxidation reactions may have been occurring in the saturated mudstone. Changes in expansion were found also to correlate strongly with changes in the ambient temperature in all specimens.

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1. Introduction

Pyrite is an iron sulphide mineral that is a commonly-occurring but minor constituent of sedimentary rocks, particularly mudrocks and shales. The mineral occurs in two main forms; cubic crystals that are typically 0.1–5 mm across and framboids that comprise spherical clusters of minute grains are typically 1–10 μm across. Cubic pyrite, which is colloquially known as fool's gold, is relatively stable in weathering environments, but on the other hand, framboidal pyrite can undergo rapid oxidation in the presence of oxygen and water. Pyrite oxidation is primarily controlled by its surface area; the reactivity increases as the grain size decreases and the surface area increases (Cripps et al., 1993). Acidity liberated by this process reacts with other rock forming minerals or construction materials containing calcium to form gypsum or other secondary minerals, including the iron oxy-hydroxide mineral, limonite. It is likely that pyrite oxidation and gypsum precipitation occur in different environments so there is unlikely to be direct replacement of pyrite with gypsum, but because the unit weight of gypsum is about half that of pyrite there is a net volume increase. In addition, the formation of gypsum crystals on discontinuities within particles and in the void space between particles results in expansion of the fill. Because the enlargement of existing crystals entails less surface energy expenditure than the precipitation of new ones, expansion can occur even if there is void space available within the fill and there is a confining pressure.

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Thus, under certain environmental conditions materials containing pyrite are prone to expansion and breakdown over time. In confined conditions such as in underfloor fill material and within hardened concrete, this expansion can lead to significant structural problems, including cracking and heaving of floor slabs. As deterioration of the rock particles in the fill continues, the fractures in the fragments become wider and more numerous, exposing more pyrite and facilitating further oxidation (Evangelou and Zhang, 1995). In effect the reaction becomes a continuous process. Some sources predict that the reaction may continue for more than 40 years (CTQ-M200, 2001; Parfitt et al., 2011), depending upon the amount of pyrite present and the rate at which the oxidation occurs.

Structures have experienced pyritic heave in many parts of the world, including Saudi Arabia, Canada, Australia, Oman, UK, USA, Japan, Norway, and Ireland. Some of these problems have been isolated events, such as the Johnson City Public Library in Tennessee, which experienced remarkable pyritic heave of the library floor (Belgeri and Siegel, 1998). Others have had a more widespread impact; for example approximately 1000 homes in Japan were damaged by pyritic heave in 2002 (Yamanaka et al., 2002) and in Canada on-going pyritic heave issues have already impacted upon an estimated 10,000 buildings. In the east of Ireland, it is estimated that between 30,000 to 50,000 homes, most of which were constructed between 1997 and 2007, during the 'Celtic Tiger' economic boom years, may be vulnerable to pyritic heave and associated structural problems.

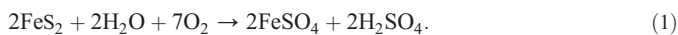
It is very difficult to predict the amount of swelling that will occur in a particular situation. Although maximum volume increase will be

limited by the amount of pyrite present, other factors are influential including, as further explained below, the temperature, as well as the amount of calcium carbonate available and the groundwater conditions. Even though pyrite-induced heave has occurred in materials containing as little as 0.1% pyrite (Penner et al., 1972), experience has shown that several years of expansion may be required for damage to manifest itself. Notable damage typically occurs after 3–5 years in Ireland and 10–15 years in Canada (CTQ-M200, 2001). Several factors have been identified to affect the rate and extent of the expansion, such as depth of the backfill, percentage of pyrite present, density of the fill, water content, position of the water table, grading of the fill, lithology of the rock, and temperature (e.g. Song and Zhang, 2009).

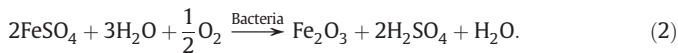
Even though the scale of the pyrite problem in the greater Dublin area is such that the authors anticipate the cost of remediation to run to several billion Euros, very little experimental work has been carried out to investigate this problem. Maher et al. (2011) report on a laboratory experiment intended to establish the susceptibility to expansion of an Irish crushed mudstone containing pyrite. The aim of the study, carried out on 600 mm diameter and 300 mm high samples of compacted material, was to develop an accelerated swell test and it was carried out using a number of potentially accelerating factors, including elevated temperatures. However, it was limited in the extent of measuring factors that influence expansion. On the other hand, this paper describes a laboratory study in which the environment of the underfloor fill was simulated. The expansion of ten specimens of the same material was monitored, with measurements of pH to confirm the occurrence of the relevant chemical reactions and consideration of the effect of sample height, water supply and temperature variation within a range typical of the Dublin area.

2. The pyrite reaction

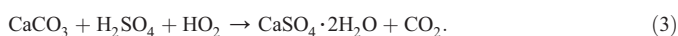
It is generally acknowledged that pyritic heave in confined fill is the consequence of three reactions, simplified versions of which are presented below. Firstly, the pyrite mineral undergoes oxidation of the sulphide, producing sulphuric acid.



In order for this reaction to take place the pyrite mineral must have contact with oxygen and water. Therefore pyritic rock only begins to react when it is exposed to the environment through quarrying or mining. The sulphuric acid produced in reaction (1) will prevent the iron sulphate undergoing further oxidation, as iron sulphate is stable in an acidic environment; however bacteria such as *Thiobacillus ferrooxidans* are capable of converting Fe(II) to Fe(III) in an acidic environment (Hawkins and Pinches, 1997), which results in the formation of the ferric iron oxide, Fe_2O_3 together with sulphuric acid:



When calcium carbonate is present, reaction with sulphuric acid produced in Eqs. (1) and (2) results in the formation of gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) and carbon dioxide (CO_2) gas.



The evolution of carbon dioxide gas, which is heavier than air, gives rise to an asphyxiation hazard in confined spaces and excavations. As is the case with most chemical and microbial reactions, the rate of oxidation of pyrite is temperature-dependent (Elberling et al., 2000), although it is generally considered a secondary influence on the reaction. In a remote site in the Arctic, permafrost conditions were utilised to freeze and encapsulate pyritic mine tailings which provided a solution to acid

mine drainage problems (Dawson and Morin, 1996; Elberling, 2005). Studies by Ahonen and Tuovinen (1992) showed an almost complete absence of microbial oxidation of Fe(II) at temperatures below 3 °C. Meldrum et al. (2001) reported oxidation of pyrite-bearing mine tailings at –2 °C, but rates were undetectable at –10 °C. It is commonly accepted that Fe(II) oxidising bacteria such as *Thiobacillus ferrooxidans* thrive best at moderate temperatures. Shafikh and Ade (2010) suggest that 36 °C is the optimum temperature for them; however this is the subject of some debate, as Hawkins and Pinches (1987) suggest 40 °C, Nixon (1978) suggests 30–35 °C and Vishniac (1974) suggests 15–20 °C. As the chemical oxidation of pyrite is an exothermic reaction, it tends to create conditions favourable to the metabolism of the bacteria.

The amount of pyrite and its form in the rock, presence of moisture, temperature and the movement of water through the fill all exert some control over rate of and maximum amount of gypsum precipitation. A supply of well oxygenated water to a fractured mudstone which contains pyrite in a framboidal form is liable to lead to a high production of gypsum. The precipitation of this gypsum depends on the solution becoming over-saturated. It is likely that this occurs towards the top of the capillary zone in the fill. In turn the location of gypsum precipitation depends on the grading of the fill and the local groundwater conditions as well as the temperature. The amount of swelling is highly sensitive to subtle changes in the character of the materials and of the environmental conditions. For example, if sulphate derived from a large volume of pyrite bearing mudstone is precipitated in a small volume of fill, then the swelling could be much greater than that produced by the amount implied by the local pyrite content. An additional cause of variation is the habit or crystal shapes developed in the gypsum. Under certain conditions, needle shaped or prismatic crystals that are orientated at right angles to the direction of stress or the surfaces of a crack or cavity in the rock and which incompletely fill the void, are formed. These will cause a much greater swelling than the same amount of tabular gypsum crystals orientated parallel to joints or cracks in the rock.

3. Experimental testing

3.1. Laboratory experiment

The laboratory experiment was designed to measure the amount/rate of expansion in simulated in situ conditions. A schematic of the apparatus developed for this purpose is shown in Fig. 1a and a photograph of all ten tests in progress is shown in Fig. 1b. As indicated, the mudstone was compacted into 229 mm diameter 6.5 mm thick medium density polyethylene tubes with lengths chosen to accommodate sample heights (H) of 500, 750 and 1000 mm (Table 1), where H was defined as the distance from the top of the water to the top of the fill. The tubes were perforated with 10 mm diameter holes that were drilled 150 mm from the lower end, to facilitate access for water, and 150 mm from the upper end to allow access to air.

Pyritic infill material was sourced from beneath the floor slab of a 5-year-old dwelling in north Co. Dublin, Ireland. This material was removed during the remediation of the house which was damaged by expanding pyritic fill material, which in common with other properties in the development, was between 500 mm and 750 mm deep. A maximum floor heave of between 6 mm and 11 mm had been noted for this and similar homes, which had resulted in widespread cracking of the walls and the doors on the ground floor were jamming. However, there was no recorded acidic attack or breakdown in the concrete below floor level. The material was described as a silty sandy fine gravel with angular and sub-angular mainly tabular or platy shaped particles of calcareous mudstone and argillaceous limestone ranging up to about 60 mm across. Further data about the chemical and mineralogical characteristics of the fill are given in Section 3.2.

The fill material was manually compacted in layers approximately 200 mm thick using a 200 mm diameter flat based tamper weighing 7 kg which, to ensure the material was not crushed and remained

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