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Concrete Deterioration and Floor Heave Due to Biogeochemical Weathering of Underlying Shale

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When the basement floor of a church in the New Edinburgh area of Ottawa heaved, it was discovered that the concrete under the floor had been reduced to a mushy consistency due to attack by sulfate solution formed by the oxidation of pyrite in the underlying shale. Gypsum was the main product in the weathered zone of the shale. It is shown that heaving resulted from the growth of gypsum crystals between the lamellae in the shale. Jarosite, which is formed by a series of oxidation reactions from pyrite, was also found in the weathered shale. Some oxidation reactions occur only in the presence of sulfur bacteria. The cementitious portion of the concrete was leached out by acid, leaving a weak porous material. The importance of safeguarding concrete in contact with potentially expansive shales from sulfate attack is emphasized.

A la suite du soulèvement d'un plancher de sous-sol dans une église du quartier New Edinburgh à Ottawa, on a constaté que le béton à la base du plancher avait acquis une consistance spongieuse à la suite de son attaque par une solution de sulfate produite par l'oxydation de la pyrite dans le schiste argileux sous-jacent. Du gypse était le principal élément dans la zone altérée du schiste argileux. On montre que le soulèvement est le résultat de la croissance de cristaux de gypse entre les lamelles du schiste argileux. On a également trouvé, dans le schiste argileux altéré, de la jarosite, résultat d'une série de réactions d'oxydations dans la pyrite. Certaines réactions d'oxydation ne se produisent qu'en présence de bactéries de soufre. Le liant de ciment à été extrait du béton par lessivage par des acides, laissant un matériau poreux peu résistant. On souligne l'importance de protéger le béton contre l'attaque des sulfates lorsqu'il se trouve en contact avec des schistes argileux susceptibles d'expansion. [Traduit par la Revue]

When extensive heaving of the basement floor of St. Luke church in the New Edinburgh area of Ottawa was reported it was suspected that it was due to expansion of pyritiferous shale. Similar occurrences had already caused problems in other buildings in the Ottawa area (Penner *et al.* 1973; Quigley *et al.* 1973). A test pit confirmed this hypothesis. The building is underlain by a black carbonaceous shale, part of the Eastview Formation. It contains about 5% organic matter (C), 8.2% calcite ($CaCO_3$), and 4.25% pyrite (FeS_2); the remainder of the rock is made up of quartz, clay minerals, and other silicates.

The church was constructed in 1913, but after 15 to 20 years the basement floor had to be repaired and it was covered with a new layer of concrete at that time. Figure 1 shows a section through the floor into the underlying shale. At the top is a 3/4-in. (1.9-cm) layer of bitumen applied about 20 years ago; below this is a layer of good concrete from 1.5 to 4 in. (3.8 to 10 cm) thick; this in turn is underlain by about 5 in. (12.7 cm) of the original concrete, now of a mushy consistency, lying directly on the weathered black shale.

The uneven thickness of the top coat of concrete suggests that the original floor had already heaved before the top layer was emplaced. When the floor was examined early in 1974 upward movement was estimated to be 2.5 in. (6.3 cm). As with most heaved floors examined by the authors the worst heave appeared to be confined to a few areas.

The top part of the shale was badly weathered (Fig. 1), but below about 1.5 ft (3.8 cm) it was competent. Numerous narrow veinlets of calcite 0.5 to 2 mm wide transect the shale. The occurrence of calcite veinlets is significant because they provide an additional source of calcium for the formation of gypsum by reaction with the sulfuric acid generated by the oxidation of pyrite (Penner *et al.* 1973). In contrast with the weathered shale found under the floor of the Bell Canada building, in which yellow jarosite ($KFe_3(SO_4)_2(OH)_6$) was plentiful, only small patches of jarosite were observed under the floor of the church. Gypsum

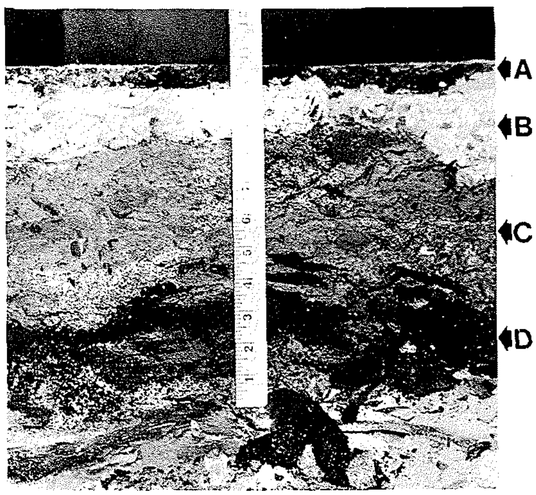

FIG. 1. Section through floor into underlying shale (scale in inches, 1 in. = 2.5 cm). (A) Tiles and bitumen; (B) toplayer of concrete; (C) mushy concrete; (D) weathered shale.

was plentiful in the weathered shale and was even found along joint and bedding planes in competent unweathered shale 3 ft (1 m) below the floor. This raises the question of how all this gypsum was formed, for it does not occur to any significant extent in the soils of the Ottawa region nor is it found in unaltered shales.

During the construction of the Ontario Children's Hospital on Smyth Road, Ottawa, detailed examination was made of freshly excavated shale underlying the foundations. Neither gypsum nor jarosite was found. Through the cooperation of the architect, geotechnical consultants, and the hospital administration, preventive measures taken to eliminate heave of the floor were omitted in a small area made accessible through a manhole in the floor. One year after construction both gypsum and jarosite have been found in the shale. During the course of a year the pyrite has evidently oxidized to sulfuric acid, and this has reacted with calcite to produce gypsum.

$$[1]\quad CaCO_3 + H_2SO_4 + H_2O \rightarrow CaSO_4 \cdot 2H_2O + CO_2$$

The exact mechanism by which pyrite converts to jarosite is uncertain but it may take place by the following series of reactions (Haldane *et al.* 1970; Ivarson 1973):

$$[2]\quad 2FeS_2 + 2H_2O + 7O_2 \rightarrow 2FeSO_4 + 2H_2SO_4$$

$$[3]\quad 4FeSO_4 + O_2 + 2H_2SO_4 \rightarrow 2Fe_2(SO_4)_3 + 2H_2O$$

$$[4]\quad 2Fe_2(SO_4)_3 + 2FeS_2 \rightarrow 6FeSO_4 + 4S$$

$$[5]\quad 3Fe_2(SO_4)_3 + 12H_2O \rightarrow 2HFe_3(SO_4)_2 \cdot (OH)_6 + 5H_2SO_4$$

Equation [4] is a possible side reaction but no sulfur was found in the altered shale.

Base exchange in an acid environment probably leads to the formation of *potassium ammonium jarosite*, the form usually found in altered shales, from the initially formed variety $HFe_3(SO_4)_2(OH)_6$.

Reaction [2] is the natural oxidation of pyrite in the presence of moist air. Reactions [3] to [4] occur at an appreciable rate only in the presence of bacteria *Thiobacillus ferro-oxidans* (Temple and Delchamps 1953). These authors also demonstrated that the grain size of pyrite is an important factor; no bacterial action was found until it had been finely ground.

The pyrite in the shale under St. Luke church is very fine grained, occurring in three distinct forms:

(1) Octahedra and combination cube and octahedra in the 3 to 10 μm range (Fig. 2a).

(2) Circular bodies 10 to 20 μm in diameter that appear under the optical microscope to have a framboidal texture but prove, when viewed with the scanning electron microscope, to consist of rather irregular circular masses (Fig. 2b), possibly, but by no means certainly, replacing micro-fossils (Ramdohr 1969).

(3) Fine-grained pyrite replacing recognizable fossils (Fig. 2c). The fine size of the pyrite in the shale undoubtedly contributes to its reactivity.

From the point of view of heaving, the important oxidation product of pyrite is sulfuric acid. Bacteria of the *Ferrobacillus–Thiobacillus* group thrive in acid environments, and once acid has been produced by the initial oxidation of pyrite (according to Eq. [2]) bacterial activity will increase and the production of sulfuric acid will be accelerated by a chain reaction mechanism. The acid produced can be diluted considerably and still be strong enough to react with calcite to produce gypsum. The acid, which is left over after the reaction with gypsum, builds up in the shale to lower the pH to the observed value of 3.

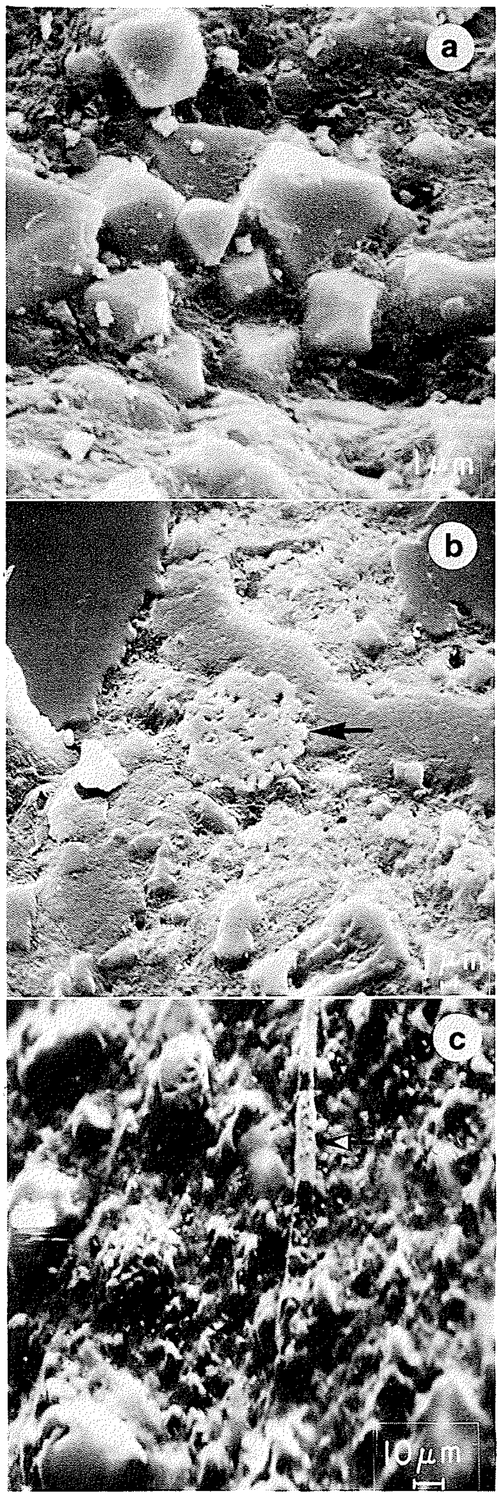

FIG. 2. Pyrite: (a) octahedra and combination cube and octahedra; (b) circular body indicated by arrow; (c) pyrite replacing fossil.

Mechanism of Expansion of Shale

Gypsum was found to form in two morphologies:

(1) bundles of fibres (Fig. 3a) growing normal to the laminations of the shale,

(2) flat, blade-shaped crystals growing parallel to the laminations (Fig. 3b).

Examination of large quantities of heaved shale has shown that the major cause of heave is the growth of the bundles of acicular crystals normal to the laminations. There is no simple explanation of why some of the gypsum forms relatively large, blade-like crystals while most

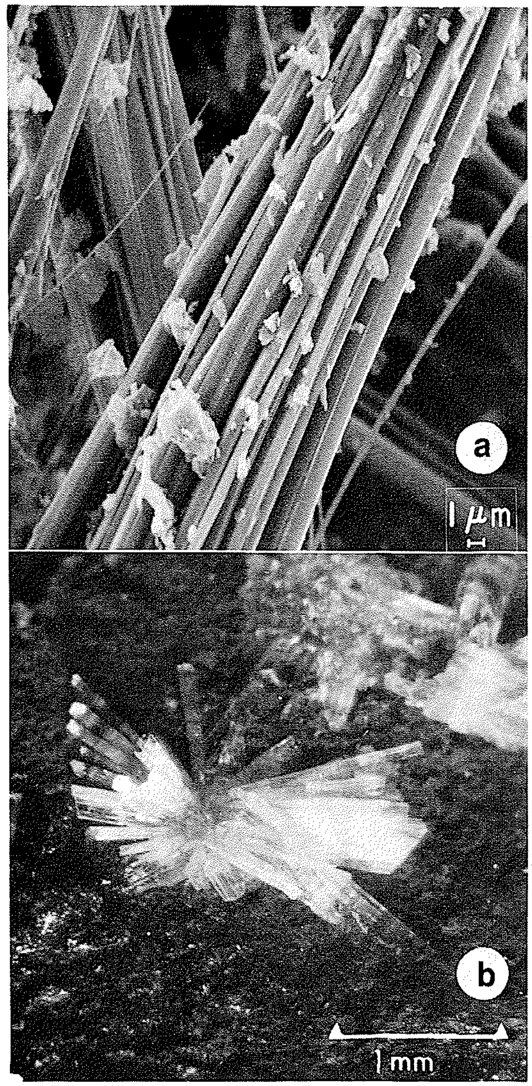

FIG. 3. (a) Bundle of acicular gypsum crystals. (b) Radiating blade-like crystals of gypsum.

of it forms fine acicular crystals. The blade-like crystals are generally found on shale that is also coated with jarosite and other ferrous and ferric sulfates. It is thought that the presence of iron salts may modify the morphology of the gypsum crystals. McCartney and Alexander (1958), Eipeltauer (1958, 1959) and Ridge and Surkevicius (1960) demonstrated that the morphology of gypsum is altered by various additives, but experiments by one of the present authors (while modifying the morphology) have failed to produce morphologies similar to those in the shale. It is thought that the physical conditions present during crystallization may also have some effect on morphology.

Conversion of calcite to gypsum does not necessarily occur at the site where the gypsum crystallizes. Acid reacts with calcite to produce a calcium sulfate solution that may be transported by capillary action to the site of crystallization.

When crystallizing under pressure, growth takes place only on a narrow rim of the concave base of the crystal (Becker and Day 1905). Because addition of new material is limited to a small section of the base, the crystal is forced upwards to produce a needle-like morphology. As crystallization proceeds, solution is brought in from surrounding areas by capillary action and the crystal slowly pushes itself up and forces apart the layers of

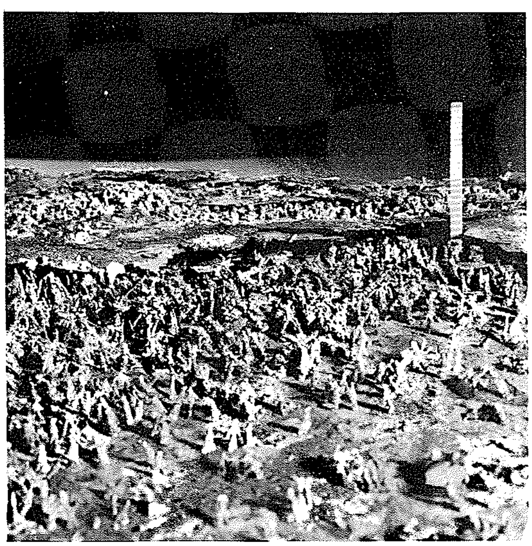

FIG. 4. Laminations in the shale forced apart by the growth of gypsum crystals (scale in millimetres, 1 mm = 0.04 in.).

the shale (Fig. 4). This process is analogous to heaving of soil by frost action from ice lensing. The occurrence of only a small amount of jarosite in the shale under the floor of the church indicates that most of the heave was due to crystallization of gypsum.

The migration of gypsum from surrounding areas towards the sites where crystallization occurred initially tends to localize the areas of maximum heave. Once cracks had developed in the floor, the pressure on the top layers of the underlying shale would be reduced, creating a pressure gradient that would tend to enhance the movement of sulfate solutions towards cracked zones where reduced solubility would cause the gypsum to crystallize.

Deterioration of Concrete

The basement floor of the church was placed in two layers, the second after an interval of about 15 years. Although the original concrete has disintegrated to a mushy consistency, the second layer is in satisfactory condition (Fig. 1). Examination with a binocular microscope of the old concrete, which lies directly on the weathered shale, showed that the cement has been largely removed, leaving large voids between the aggregate particles. When subjected to acid attack, it is usual for the soluble phases of cement, calcium hydroxide and calcium silicate hydrate to be removed, leaving a weak porous material such as that observed in this concrete.

Gypsum was detected as a coating on occasional aggregate grains in the original concrete. In contrast with the gypsum in the shale (occurring as elongated crystals growing parallel to the *c* axis), that in the concrete has formed tabular crystals which grow parallel to the *b* axis (010) (Figs. 5b and 6b). These tabular crystals consist of a pile of flat platy crystals (Fig. 6b).

Fine needle-like crystals filled some of the voids in the concrete (Fig. 7a); an enlarged view is show in the micrograph (Fig. 7b) in which rhombohedral terminations characteristic of calcite can be seen (this identification was confirmed by X-ray diffraction). Calcite is usually a major constituent of old concrete, but because of acid attack it has largely been removed from the sample. Needle-like calcite crystals fill large voids, suggesting that calcite is a secondary deposit emplaced after the

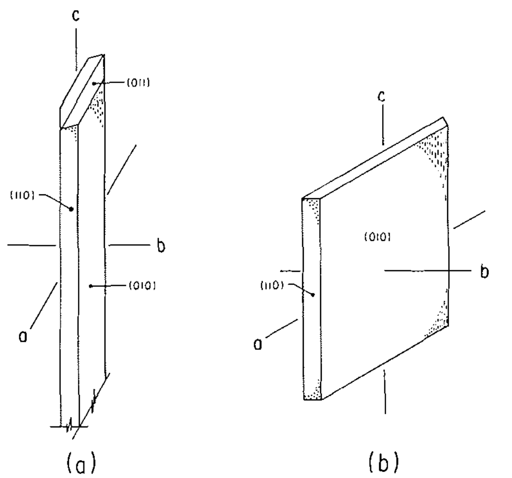

FIG. 5. (a) Acicular gypsum crystal elongated parallel to *c* axis. (b) Tabular gypsum crystal with large (010) face.

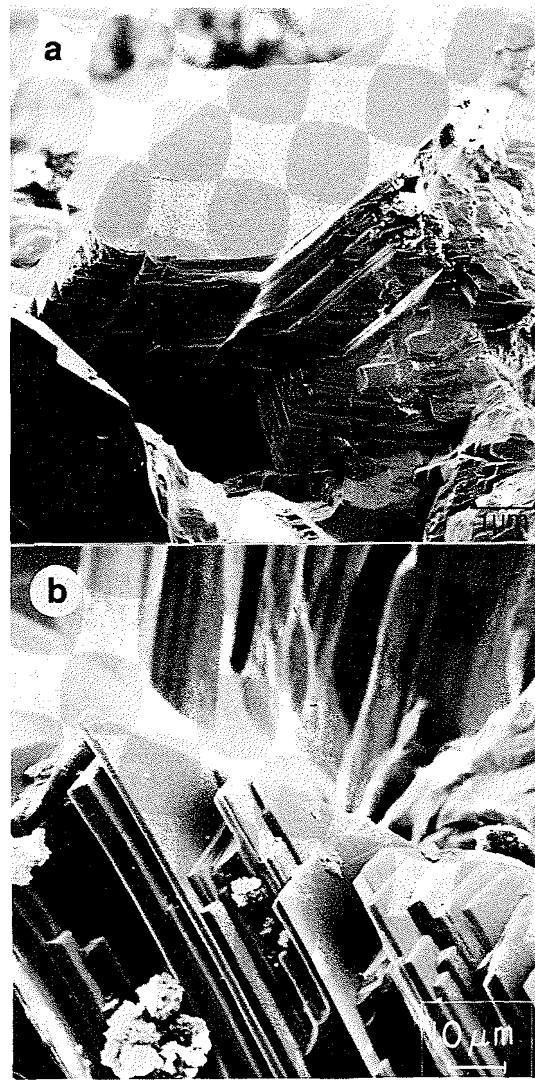

FIG. 6. (a) Tabular gypsum crystal. (b) Enlarged view of (a) showing stacking of platy crystals comprising tabular crystal.

original cementitious constituents were removed.

Practical Implications of Disintegration of Concrete in Contact with Carbonaceous Pyritic Shale

The basement floor of the church is the only confirmed case of disintegration of concrete in contact with pyrite-rich shale known to the authors in the Ottawa area. Another case of weak concrete was observed in the basement floor of a house laid over rock fill containing pyritic shale, but the method of construction might have accounted for the poor concrete. Sulfate attack on concrete in contact with alum shales has been documented in Oslo, Norway, by Moum and Rosenquist (1960).

In the Bell Canada building (Penner *et al.* 1970) the footings for the columns were set in shale and surrounded by shale in the process of weathering. Examination proved that the concrete in these footings was sound. It might be useful to consider why the concrete floor of the church basement was destroyed while the footings in the Bell building were not.

It is widely held that sulfate attack occurs only when a sulfate solution is drawn through relatively impervious concrete by evaporation from one side of a slab or when the slab is immersed in the solution. The bases of the columns in the Bell building are 1.2 m below the floor, well above the water table which is more than 3 m down. Gypsum was almost absent from the shale under the column footings. For reasons already discussed, the sulfate solutions would tend to be drawn away from such areas to those where heave was taking place so that it would be unlikely that the concrete of the columns would be subjected to sulfate attack.

The possibility of decomposition of concrete in contact with heaving shale indicates that care should be taken to minimize all contact between shale and concrete in buildings con-

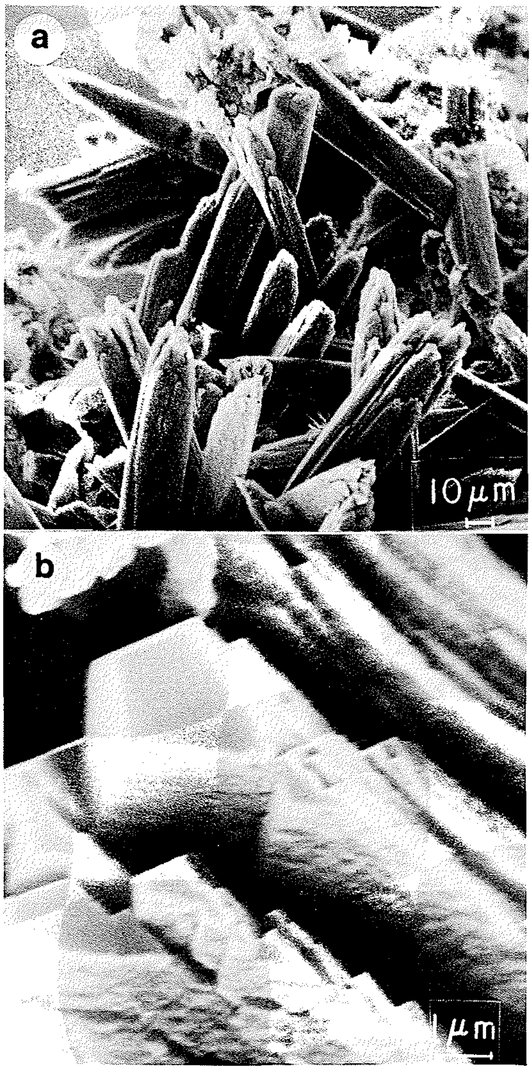

FIG. 7. (a) Calcite crystals. (b) Enlarged view showing rhombohedral terminations.

structed on such formations. In this context the preventive measure of coating freshly excavated shale with concrete to try to exclude air and therefore prevent oxidation of the pyrite in the shale may be hazardous if it is not successful.

Conclusions

The basement floor of a church in the New Edinburgh area of Ottawa heaved as a result of expansion of the underlying black pyritic shale of the Eastview Formation. Expansion was largely the result of the growth of acicular gypsum crystals between partings in the shale. Both the small amount of jarosite found and sulfuric acid are products of the oxidation of pyrite in the presence of bacteria. Gypsum was formed by the reaction of sulfuric acid with calcite.

The concrete of the church basement floor deteriorated to a mushy consistency as a result of attack from sulfate solutions generated by the oxidation of pyrite in the shale.

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