# **Expansive Pyritic Shales**

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#### ABSTRACT

Although much attention has been directed to the solution of problems created by swelling soils, relatively little effort has been devoted to the solution of problems created by expanding pyritic shales. This is a very real problem that has caused extensive damage to many structures in the United States and abroad. In an effort to call attention to this problem and to the need for further research directed at its solution, the current state of the art with respect to swelling pyritic shales is presented and discussed. The factors and mechanisms responsible for swelling of pyritic shales are identified; methods of testing and evaluating the potentially expensive shales are outlined; and ways of controlling or eliminating the problem are summarized. A number of case histories involving experiences with expansive pyritic shales are included to illustrate the potential seriousness of the problem and to underscore the need for additional research.

It has been reported that the average annual losses in this century from floods, earthquakes, hurricanes, and tornadoes have been less than one-half of the damages from expansive soils (1). Geotechnical engineers who deal with expansive soils or shales have been faced with the decision of either accepting a certain risk in view of the uncertain response of foundation materials to changing environmental conditions or choosing safe but generally expensive foundation systems. There has been a general reluctance to adopt the latter approach because it has appeared that strengthening the foundation can sometimes cost more than correcting the damages that can result from foundation heaving. It is important to note, however, that whereas considerable money has been spent to control floods, relatively little has been invested to develop and implement methods for controlling or mitigating damages associated with foundation heaving.

A family of expansive materials that has caused significant damage to many structures, but has been widely overlooked, is pyritic shale. In the past damage to structures by expanding pyritic shale was not readily identified. Consequently, damages were often attributed to differential settlement, frost heave, subsidence, or poor construction practices. More recently, however, the problems associated with expansive pyritic shales have been better recognized. Spanovich (2) suggests that part of the reason for an increased frequency of problems caused by expanding shale is a result of advances in powerful excavation equipment that can produce greater cuts and expose deeper fresh shale strata, and also as a result of the increased use of slab-on-grade construction (because of economic constraints) without providing crawl spaces to absorb heave.

Shale is a sedimentary rock formed by the compaction and cementation processes acting on clay, silt, and sand particles. Compaction shale is a transition material between soil and rock susceptible to significant weathering and slaking, whereas cemented

shale exhibits the general characteristics of sounder rock ( $\underline{3}$ ). Because of its abundance (it constitutes about 50 percent of all exposed rock), shale is often used as an engineering material to build fills and embankments, highway bases in certain cases, and as a foundation for all types of structures, including bridges and other transportation structures. Extensive studies have been conducted on the evaluation of many shales for highway use ( $\underline{4-9}$ ), but little attention has been directed in these studies to pyritic shales either as a construction or foundation material.

The importance of understanding the nature of pyritic shale is two-fold. First, it is an expansive material for which the mechanism of expansion or swelling is different from what is known for ordinary shales or soils (10). The swelling phenomena observed in pyritic shales are primarily caused by the volume changes induced by the chemical alteration of sulfide minerals. Second, lechate from pyritic shale can attack concrete and cause deterioration. This phenomenon is known as "sulfate attack" (11) and also results from the chemical alteration of minerals, this time leading to volume changes in concrete.

The purpose of this paper is to summarize the existing information and knowledge on pyritic shales. Factors and mechanisms of swelling in pyritic shales and experimental methods of evaluating swelling potential are reviewed. A number of case histories related to heaving damage and concrete deterioration are presented and discussed. Finally, known methods of controlling the problem are described and research needs are identified.

## FACTORS AND MECHANISMS OF SWELLING IN PYRITIC SHALES

## Primary and Secondary Reactions

Heave observed in certain shales has been identified to be the result of the oxidation of sulfide minerals, the most common of which are pyrite and marcasite. Pyrite, or ferrous disulfide (FeS2), is a yellowish mineral with a metallic glint that long ago earned the name of "fool's gold." Pyrite is generally regarded as stable or slightly reactive under normal conditions; however, on exposure to dampness for a long period in the presence of air, it oxidizes and expands. Pyrite is widespread, being the most common iron sulfide mineral. It occurs in rocks of all types and geologic ages, most often in metamorphic and sedimentary rock. Marcasite has the same chemical formula as pyrite, but X-ray analysis reyeals a different atomic arrangement. Marcasite is generally gray in color and is more susceptible to oxidation than pyrite. However, it is less abundant than pyrite (12).

Crystallographically, the oxidation and expansion mechanisms of the sulfide minerals to sulfate are quite readily understood. The pyrite structure is stacking of close-packed hexagonal sheets of sulfide ions with iron occupying the interstices of the sulfide layer. The packing density of this configuration is related to the radius of the sulfide ion, which is 1.85 angstroms, producing a volume of 26.14 cubic angstroms. In the sulfate structure, each atom of sulfur is surrounded by four atoms of oxygen in

tetrahedral coordination. The packing density of the corresponding sulfate compounds will depend on the radius of the sulfate ion, which is 2.805 angstroms. This results in a volume of 94.4 cubic angstroms, which represents an approximate volume increase of 350 percent per packing unit  $(\underline{13})$ . If calcium is present, gypsum may be formed, which may give rise to an eight-fold increase in volume over the original sulfide  $(\underline{14})$ .

The primary oxidation reactions occurring in the sulfide alteration process are thought to be as follows (15):

$$2\text{FeS}_2 + 2\text{H}_2\text{O} + 7\text{O}_2 \rightarrow 2\text{FeSO}_4 + 2\text{H}_2\text{SO}_4$$
 (I)  
ferrous sulfuric  
sulfate acid

$$4\text{FeSO}_4 + \text{O}_2 + 2\text{H}_2\text{SO}_4 \rightarrow 2\text{Fe}_2(\text{SO}_4)_2 + 2\text{H}_2\text{O}$$
 (II) ferrous sulfate sulfate

$$7\text{Fe}_2(\text{SO}_4)_2 + \text{Fe}\text{S}_2 + 8\text{H}_2\text{O} \rightarrow 15\text{Fe}\text{SO}_4 + 8\text{H}_2\text{SO}_4$$
 (III) ferric pyrite sulfate

Reaction I will normally proceed unaided, although the oxidation of the sulfide may be assisted by autotrophic bacteria. Reaction II is thought to be entirely due to the ferrobacillus-thiobacillus bacteria, because the reaction cannot proceed in an acid environment. This was shown by Goldhaber and Reynolds (16) in a series of experiments. Data obtained on samples drawn periodically and analyzed for total sulfur in solution, thiosulfate and sulfate, indicated that the rate of oxidation increased markedly as pH increased, particularly above a pH of 7. Reaction III oxidizes more pyrite by reacting with ferric sulfate, a strong oxidizing agent produced in Reaction II.

Gypsum is known to form from the reaction of sulfuric acid with calcite. Jarosite, another main reaction product, is essentially insoluble in water and forms most readily in an acid environment  $(\underline{12})$ . Gillot et al.  $(\underline{17})$  stated that when lime is present, gypsum forms together with iron hydroxides, whereas jarosite and iron hydroxides appear to be the main reaction products when lime is absent.

Some secondary minerals are also formed from the chemical reactions (2). These minerals are (a) melanterite, an aqua to white mineral that will dissolve in groundwater; (b) rozenite, an aqua to white, stringy mineral that forms by the loss of bound water when sulfuric acid dehydrates melanterite; (c) coquimbite, composed of soluble, blue-green or white crystals that form with alternate wetting and drying of rozenite; (d) kaolinite, a white, powdery, stable clay mineral that remains in addition to jarosite; and (e) limonite, a rusty, red powder or stain that results from the further breakdown of jarosite.

The roles of the primary and secondary reaction products derived from the oxidation of sulfide minerals are somewhat controversial. This is particularly true in the case of gypsum. It is not thoroughly understood whether heave is caused by the formation of gypsum crystals or whether gypsum only exhibits a void-filling function. However, there is general agreement that the secondary minerals—jarosite, rozenite, coquimbite, kaolinite, and limonite—serve only to occupy voids formed during the expansion process.

Studies performed on black shales in Pennsylvania, West Virginia, and eastern Ohio have suggested that the expansion of pyritic shales is sometimes erroneously attributed to the formation of gypsum crystals that only inhabit voids vacated by dissolved minerals  $(\underline{17})$ . However, Quigley et al.

 $(\underline{18})$  and others  $(\underline{12},\underline{19}-\underline{21})$  state that the growth of gypsum crystals within shale is the primary cause of heave. Resolution of this issue is of practical as well as theoretical importance. If heave is caused by the formation of gypsum, black shales that are essentially free of calcite would be considered safe from an engineering standpoint because the supply of calcium ions would be limited and the amount of gypsum formed would be small.

The chemistry of the formation of gypsum in pyritic shales appears to be rather straightforward, involving an attack on calcite by sulfuric acid formed by the oxidation of pyrite. The acid reacts with calcite to produce a calcium sulfate solution that may be transported to the site of crystallization. The alteration products (gypsum) are frequently located near existing pyrite, which indicate that they precipitate out of solution and begin to grow after traversing a short diffusion path (17). As crystallization proceeds, additional solution is brought in from surrounding areas by capillary action, and the growing crystals force the shale layers apart. This process is analogous to frost heave by ice lenses.

By the use of a scanning electron microscope on samples of weathered pyritic shale, Grattan-Bellew and Eden (22) found that gypsum can form in two morphologies: (a) bundles of fibers growing normal to the bedding planes of the shale, and (b) flat, blade-shaped crystals growing parallel to the bedding planes. Examination of large quantities of heaved shale has revealed that the major cause of heave is the growth of the bundles normal to the laminations. Why some of the gypsum forms bladelike crystals while others form bundles is not fully understood. It is speculated that the presence of iron salts and the physical conditions present during the crystallization may modify the morphology of gypsum crystals.

An interesting argument for the expansion of pyritic shales caused by the growth of gypsum crystals is presented by Coveney and Parizek (23). According to these researchers, gypsum, the chief host of sulfur in the weathered rock, is considerably less dense (specific gravity = 2.36) than pyrite (specific gravity = 5.02), the chief host mineral in the unweathered rock. The sulfur-bearing minerals in the weathered rock occupy nearly twice as much volume as that in the fresh rock. Thus gypsum formed by weathering occupies about twice the volume of pyrite, causing the shale to swell roughly 1.55 percent by volume. Because the shale is effectively confined at the bottom and the sides, this would result in thickening of the shale layers by that percentage, which would yield an average floor uplift of 0.8 in. for a 4- to 5-ft-thick pyritic shale layer. This alone would be too small to account for the observed heave in many cases. However, these calculations fail to consider that the gypsum crystals occupy only a small portion of the volume of the space they form. Observations of a polished surface of a typical veinlet show that only 27 percent of the plane surface area is occupied by gypsum crystals and the remaining 73 percent consists of voids. The pore space developed by gypsum growth thus amounts to a minimum of three times the space occupied by the gypsum. Taking this into account, an overall volume increase of 8 percent can be expected. This would correspond to a heave of about 4 to 4.5 in. for the same 4- to 5-ft pyritic shale layer, which is in general agreement with heave observed at some sites. The magnitude of the uplift force created by this mineral growth has not been accurately determined, but it has been suggested that heave pressures as high as 12,000 lb/ft2 may develop (2).

Although melanterite is another possible reaction product that can contribute to the expansion phenomena in shales, it has not been possible to identify its role clearly. This is because it is unstable and reverts to an anhydrous powder form on exposure to normal room temperature and atmosphere (12). It is also extremely soluble; therefore, the usual sampling of the shale by core drilling would most likely dissolve and wash out the melanterite crystals, leaving little if any evidence that the mineral was present.

## Role of Autotrophic Bacteria

Some controversy has arisen in the discussion of whether or not autotrophic bacteria are in part responsible for the expansion of pyritic shales. It is suggested by some researchers (15,18,21) that the bacteria play the role of a catalyst in the oxidation of sulfides to sulfates. As Berard (24) points out, however, the oxidation of the sulfide minerals will proceed without any bacteria assistance, but it is certainly catalyzed by oxidizing bacteria. In the case of unstable minerals such as pyrite and pyrhotite, bacterial action is not necessary to produce rapid oxidation. Given the right oxidation potential, which is often found in soils above the water table, these two sulfides will transform into lower free energy minerals.

The origin of the autotrophic bacteria can be most easily identified by observing the geologic origin of black shales. Black shales are believed to have formed in stagnant marine environments characterized by reducing conditions in which organic mineral collected and anaerobic bacteria were active. The common bacteria present were those whose role was to reduce sulfates to sulfides in the bottom muds. These bacteria play a role in the formation of black shale, whereas the oxidizing autotrophs of the thiobacillus type are functional in the heaving process (18).

The autotrophic bacteria grow and multiply using energy from the oxidation of inorganic compounds. Thiobacilli bacteria are probably most active in the early phases of the reaction, whereas ferrobacilli play a dominant role only later when there is a reduction in pH because of the production of sulfuric acid. Sulfuric acid is formed in the oxidation reactions and produces an environment favorable to the growth of this type of bacteria. According to Penner et al. (15), these bacteria require an acid environment with an optimum pH of 2.2. They are known to go into dormancy at a pH value greater than 4.5, and optimum temperature is thought to be around 95°F.

#### Additional Factors That Influence Heaving

In addition to the previously mentioned factors, there are still other factors that merit consideration and discussion. These include groundwater and site grading.

#### Groundwater

The exact role of groundwater in the heaving process is not clearly understood. Studies have indicated that if the sulfide-bearing stratum is totally submerged, the heaving process does not occur, most likely because of the absence of necessary oxygen (21). On the other hand, the oxidation of potentially exponsive sulfide minerals will begin in an environment relatively free of water, such as that above the water table (24). Therefore, the lowering

of the groundwater level by excavation or peripheral drains around the structure may help trigger the oxidation and consequently the expansion of the sulfide-bearing stratum.

## Site Grading

The excavation of soil above strata that contain potentially expansive sulfide minerals may enhance the oxidation of sulfide minerals. First, grading may remove the weathered rock and thus expose fresh rock that contains a higher concentration of sulfide minerals (2). Second, removal of overburden pressure can cause slight rebounding and fracturing in the underlying strata, further exposing the sulfide minerals to air and moisture (25). Finally, a secondary effect of excavation is that the structure is placed closer to the sulfide layer, which may cause a loss of any insulation of the sulfide-bearing stratum from the heat of the structure. This insulation from heat given off by the structure is important. As emphasized by Berard (24), a temperature gradient is responsible for the upward migration of water and dissolved oxygen, which in turn causes the oxidation of the sulfide. In addition, Quigley and Vogan (21) suggest that a warmer, more humid environment caused by the higher temperatures is more favorable to the growth of bacteria.

#### TESTING AND EVALUATION OF SWELLING POTENTIAL

A number of testing schemes are available to evaluate the swelling potential of shales in general and pyritic shales in particular. These are the free swell test and the modified swell test  $(\underline{14},\underline{26})$ , induced gypsum crystal growth  $(\underline{20})$ , and total sulfur analysis  $(\underline{12})$ . Although the results of these tests generally give a good indication of the amount and rate of swelling expected in situ, the reliability of quantitative predictions is not well established.

#### Free Swell Test

In the free swell test the rock core specimen rests on a pedestal mounted to the base of a rigid frame, as shown in Figure 1 (26), and is permitted to expand unrestricted in all directions. The amount of swelling is measured in the vertical direction with a dial gage, which is initially zeroed against a standard stainless-steel bar. As noted earlier, the expansion of shaley rocks is significantly larger in the direction perpendicular to the bedding planes

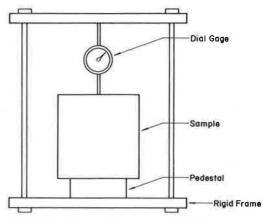


FIGURE 1 Free swell test apparatus (26).

than in a direction parallel to the layering. The testing assembly is stored in a curing room under conditions of 100 percent humidity and is removed periodically to take readings. The strains calculated from these readings can be plotted as a function of elapsed time (log scale) to get an indication of the amount of swelling expected to occur in the field  $(\underline{14})$ .

#### Modified Swell Test

The modified swell test is a semi-confined test in which the rock sample is submerged in distilled water and the strains are monitored in one direction by a dial gage, as shown in Figure 2  $(\underline{14})$ . A con-

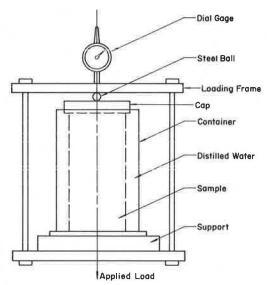


FIGURE 2 Modified swell test apparatus (14).

stant load is applied to the rock sample in the direction of measurement while deformation in other directions remains unrestricted. Again the test data can be analyzed by a semi-logarithmic plot of strain versus time. The average slope of such a plot gives an index of swelling potential, which indicates the tendency of the rock to expand on stress relief (14).

It should be noted that the modified and free swell tests are simple and inexpensive to carry out. For this reason it becomes easy to perform a large number of tests to aid in the evaluation of potential problems. The major drawback with these tests, however, is that they are time-consuming. Reactions that cause swelling generally take long time periods.

## Induced Gypsum Crystal Growth

In the induced gypsum crystal growth test a modified one-dimensional consolidometer is used to monitor heave in shale while gypsum crystal growth is induced artificially ( $\underline{20}$ ). The testing device resembles a fixed-ring consolidometer used in soil testing with the addition of two fluid inlets and reservoirs located on opposite sides. Specimens are exposed to solutions of CaCl $_2$  and (NH $_4$ )  $_2$ SO $_4$  induced simultaneously under the same head at opposite fluid inlets. The reaction to form gypsum crystals in this case is

 $CaCl_2 + (NH_4)_2 SO_4 + 2H_2 O \rightarrow CaSO_4 \cdot 2H_2 O \downarrow + 2NH_4Cl$ gypsum

The samples remain submerged during the entire period of testing, and volume change is monitored throughout the test. After completion of testing the samples are removed for photographing and are analyzed by a scanning electron microscope or by X-ray diffraction to monitor gypsum formation.

## Total Sulfur Analysis

In total sulfur analysis the total sulfur content of a sample is assumed to consist of sulfate, pyritic sulfur, and organic sulfur. The amount of sulfate sulfur represents a measure of the degree of oxidation, and thus gives an indication of the amount of expansion that has taken place in the sample before sampling. The amount of pyritic sulfur represents the amount of potential expansion that can take place when all of the sulfides completely oxidize. The organic sulfur content, which is needed to complete the measure of total sulfur, is not believed to be involved in the expansive reaction (27). Total sulfur analysis is valuable in assessing probable ultimate heave values and percent heave to which a structure has already been subjected.

## Sampling

Sampling for the tests previously described must be executed with great care. The usual types of samples tested for potentially expansive sulfide minerals are rock cores and samples removed by hand from excavations. If sampling is to be done at a site where heaving has occurred, no water should be used in the drilling operation, because the drilling water may wash out or dissolve some or all of the melanterite crystals. At sites where heaving has already occurred, test pits are a preferable method of investigation and sampling (12). Specimens for testing should be prepared for such testing as soon after sampling as possible. The rock cores or other samples should be wrapped in plastic immediately at the site to minimize changes in the natural moisture conditions.

In the case of testing the samples for autotrophic bacteria, extracts of water samples from boreholes should be incubated in a ferrous sulfate medium at a pH of 2 to 3 for 2 weeks on a rotary shaker to establish if microorganisms exist. The samples should then be examined microscopically for the presence of autotrophic bacteria of the thiobacillus ferroxidan and ferrobacillus ferroxidan types (17).

## PROBLEMS RESULTING FROM PYRITIC SHALES: CASE HISTORIES

The following case histories illustrate factors that contribute to the expansion of pyritic shales and demonstrate the potential damaging consequences of such expansion. In addition, some of the case histories presented demonstrate that the oxidation of pyritic shales can lead to reaction products that can cause damage to structures through concrete deterioration, which could perhaps be more widespread and more devastating than the heaving problem. Methods of solving the problems illustrated in the case histories will be discussed in a general manner later in the paper.

## West Virginia University Hospital and Basic Sciences Building

The investigation of the West Virginia University (WVU) Hospital and Basic Sciences Building was performed by Triad Engineering Consultants, Inc., of Morgantown, West Virginia, in September 1981 (28). The building exhibited signs of distress in the form of cracking in the non-load-bearing tile walls, along with noticeable heaving of the concrete floor slabs. Black carbonaceous shales containing pyrites were found in the rock strata below the building. An investigation was performed to see if the wall distress and floor heave could possibly be attributed to underlying expansive shales.

The distressed areas were found at various locations over the complete building site; however, the areas of greatest distress appeared to have occurred on the ground floor. Primarily, these distressed locations were most severe in areas that overlie the black shale strata located relatively near the edge of the excavation for the construction of the basement.

The evaluation of the expansive shales revealed that the black carbonaceous seam appeared to be the cause of heaving and cracking in the floors and walls. The shale seam generally lies below the ground floor elevation and above the basement floor elevation, and has a variable thickness ranging from 0.9 to 2.6 ft. When the basement area was excavated, most if not all of the black shale was removed from under the proposed basement floor. For this reason, the heave appeared only in the ground floor and not in the basement.

The total sulfur test was performed on samples obtained from areas of heave. In the black shale the percentage of total sulfur varied between 1.86 and 5.96, depending on location. The percentage of sulfate sulfur ranged from 0.03 to 1.61; however, the higher values were found on samples obtained from under areas where floor heaving and distress cracking were the severest. This conformed to previous research, which found that the expansion of the shales is caused by the reaction of the pyrites into various forms of sulfates. In general the movements occurred near the edge of the basement excavation, but just a few feet away from the edge of the excavation cut little or no movement occurred. Also, the percentage of sulfate sulfur was greater in the black shale nearest to the cut of the basement and decreased with increasing distance from the cut face.

It was suggested that the reason for increased swell next to the cut was because of the excavation. The excavation for the basement intersected the black shale seam that allowed air and aerated groundwater into the shale, which reacted with the pyrites.

The areas where expansion of the shales had already occurred had not undergone the total amount of expansion possible because considerable amounts of pyrites had not reacted. Because of the relatively short monitoring period, the amount of additional expansion could not be accurately determined; however, it appeared that the expansion at the time of testing had not yet reached 50 percent. Also, it was noted that as the heave continued the rate of expansion should increase because the rock strata becomes more fractured, which would allow access of more air and aerated water to unreacted pyrites.

#### Wheeling Hospital

Heaving of floor slabs, partitions, and column coverings was observed in portions of the basement of the Wheeling Hospital following flooding of the lower levels of the hospital on Labor Day 1975. In

addition, the elevator shaft of the hospital also appeared to have heaved upward as observed by the cracking of the floor slabs adjacent to the elevator shaft in the upper floors.

The initial foundation investigation report for the hospital, which was completed in July 1970, indicated the presence of potentially expansive pyritic shales within the foundation and recommended protective measures to be taken to reduce or eliminate expansion. A later investigation in June 1980 reported the presence of these expansive shales and their reaction products beneath one of the most badly heaved areas of the basement floor slab. Later in 1980 a rather extensive set of elevation readings was taken on the floor slab, and heave monitoring positions were established to permit more precise measurements of heave to be made on a regular basis.

In June 1982 Triad Engineering Consultants, Inc. (27), was contacted to perform an independent investigation of the situation to verify the presence and areal extent of the expansive shale problem, to determine the potential long-term effects of the expansive shales in terms of how much additional movement can be expected and over what time period, and to suggest possible remedial measures to solve the problem. The investigation included damage inspection, borings, examination and testing (by total sulfur analysis) of soil and shale samples obtained from the distressed areas, and the correlation of the test results with heave measurements to predict the magnitude and rate of future heaving.

The foundation material was found to consist of a heterogenous fill containing silty clay, sand, gravel, pyritic shale, and coal fragments underlain by pyritic gray shales up to a depth of 11.5 ft below the basement level. No groundwater was encountered. Sulfur content tests indicated that total sulfur percentages varied from a low of 1.49 to a high of 4.84 in the gray shale. The sulfate sulfur contents varied between 0.01 and 2.70 percent, with higher values generally indicating good correlation with the amount of heaving observed. It was concluded that heaving was a direct result of expansion caused by the oxidation of sulfide minerals in the underlying soil and rock.

Based on the elevation readings provided, use of linear time-heave curves, and an average volume expansion factor calculated by dividing the apparent heave by the volume of sulfates formed to date, it was estimated that up to approximately 4 in. of heave had occurred through July 1982. The future or remaining heave was estimated as the product of the same expansion factor and the volume of the unreacted pyrites in the underlying strata; it was projected to be as much as 11 in.

In this instance the pyritic shales causing heaving of the basement floor were found to be mostly close to the surface, and therefore the most effective and economical type of remedial construction would involve the removal of the existing floor slabs, the excavation of the pyritic shales, backfill with well-compacted granular material, and the construction of new floor slabs. The only reasonable solution to the heaving problem for the elevator shaft appeared to be the installation of a system of rock anchors to prevent future heaving. However, the expected rates of heaving were relatively slow. Consequently, although these remedial measures will eventually be necessary, it was not recommended that they be undertaken immediately.

## WVU Engineering Sciences Building

The Engineering Sciences Building consists of a 10-story tower section surrounded on three sides by

low-rise structures. Shortly after the building was constructed in 1959, some minor cracking and distress were observed in the floor slabs and non-load-bearing walls. Movements have continued until the present time, and maintenance has been performed periodically. Detailed maintenance records were not kept; however, it appears that expensive repairs were made twice and possibly three times during the 1960s. Since the summer of 1971, movements apparently accelerated according to maintenance personnel.

Level surveys of the building were conducted to determine the vertical direction of movement (i.e., heave or settlement) and to obtain an approximate measure of the amount of movement. The survey indicated that little or no movement has occurred under the tower section of the building. The section of the building where the basement floor is supported on grade had experienced the largest movements and the heaviest damage. Slabs on grade have heaved up to 2.5 in. and column foundations have heaved up to 3.0 in

A subsurface investigation was conducted to determine the nature of the material causing the heave. Pyrites were encountered in all of the borings drilled as part of the geotechnical investigation. Borings and test pits, made from the level of the basement floor, found such sulfate minerals as gypsum and jarosite crystals and limonite staining on the bedding planes, all evidence of previous expansion of sulfide minerals.

Chemical analyses were performed on several of the rock samples to determine the sulfur content. It is suggested that a sulfide content as low as 0.5 percent is potentially expansive, and a sulfide content of greater than 1 or 2 percent is considered highly expansive (25). The test results indicated that the rock had high expansion potential as evidenced by both the total sulfur and the sulfide sulfur contents.

It appeared that the excavation for the building removed 30 to 50 ft of overburden soil, which allowed the joints and bedding planes to open slightly. The bedding planes are nearly horizontal, and the rock is relatively impervious in the direction perpendicular to the bedding. Therefore vertical penetration of air and moisture was restricted. Thus only surficial expansion of the rock, which is relatively small, was possible in the subbasement of the building where vertical penetration of air and moisture took place. This was not the case for the rock behind the retaining walls separating the subbasement and basement sections. Here the bedding planes are exposed to lateral infiltration of air and oxygen-rich water, which migrate horizontally with little resistance, including the chemical reaction leading to heave. As a result, approximately 7 to 10 ft of rock below the basement floor near the retaining walls is participating in the expansion, whereas probably only 2 or 3 ft of rock is participating in the expansion below the subbasement. Consequently, the heave is maximum near the retaining walls and decreases with increasing distance from the walls. In addition, it was found that, considering the sulfur content of the rocks below the Engineering Building and the thickness of the rock that might be involved in the heave, it is reasonable to expect that the ultimate heave might be twice the present magnitude.

A program of remedial construction designed to correct these problems will be undertaken in the near future. The heaved floor slabs will be removed and replaced with a structural floor system supported on columns founded below the expansive strata. Those column footings that have heaved will

be tied down with rock anchors to prevent future heaving.

#### Three-Story Steel Frame Building in Pittsburgh

A three-story steel frame building in Pittsburgh (29) with masonry exterior was built on natural soil. Heavy concentrated column loads were supported by spread footings bearing the bedrock. The upper surface of bedrock was a thin layer of black carbonaceous shale about 18 in. thick. Beneath this was a layer of limestone 3 to 4 ft thick, and the next 4 ft were gray weathered clay shale interbedded with limestone.

The first portion of the building was built in 1931, and subsequently several additions were constructed. Crack damage was first reported in 1950 in the first floor walls and slabs. The damage spread through the second and third floors and increased slowly at a uniform rate. The concrete floor of the gymnasium heaved in 1954 and was 6 in. higher in the center than the perimeter. The floor was replaced in 1956, and during the next 15 years the gymnasium floor again developed a differential movement of 4 in.

Two test borings were drilled inside the building. The shale beneath the floor slabs and footings was examined petrographically and chemically. Iron sulfides and their alteration products from the oxidation of the sulfides were found in various mineral forms. It was concluded that air or moisture or both caused the alteration and expansion. Heat from a utility tunnel beneath the floor slabs around the perimeter of the building probably accelerated the reaction. The amount of sulfur and sulfate in the altered shale compared with the unaltered sample indicated that the oxidation would continue and that the amount of total expansion would be twice the amount that had already occurred.

# Black Shale Heaving and Concrete Deterioration in Ottawa, Ontario, Canada

The Therapy Treatment Building of the Rideau Health and Occupational Center located in southeast Ottawa is a two-story building with basement areas, an area without basement, and a deep swimming pool, all founded on shale bedrock. The two-story portion without the basement and founded on intact rock experienced heave, whereas the basement area and the swimming pool were not affected. The concrete floor slab heaved up 3 in. between 1965 and 1969. The floor of the second story auditorium was also considerably warped, indicating that the interior columns also had heaved.

A concrete heating and service tunnel was constructed in a trench dug into rock around the perimeter of the two-story building. The central mass of the shale bedrock was not excavated and forms a horizontal rock plateau 6 to 7 ft above the bottom of the service tunnel. The interior columns of the building are founded on this rock plateau and are lightly loaded, as they support only the weight of the second floor auditorium. The ground floor slab consists of concrete poured over about 18 in. of granular material on top of the rock plateau. During the winter the service tunnels are hot (about 85°F). During the summer, when the heat is off, the rock is probably close to ambient rock temperature.

The important heave mechanism at this site was believed to be geochemical alteration of sulfides in the shale bedrock to secondary sulfates having a much greater volume. The amount of secondary gypsum

found in the core samples was noted to correspond to the amount of heave.

Quigley and Vogan (21) presented the following explanation for the secondary gypsum growth related to this case.

- 1. Secondary gypsum occurred above the water table in a zone that was probably partially saturated by capillary rise. The environment was humid because of the heat from the service tunnel, which was ideal for the growth of aerobic, oxidizing bacteria, which were confirmed to exist from cultures grown in the laboratory.
- 2. Chemical analysis confirmed the presence of sulfur in the black shale (as pyrite).
- 3. Autotrophic bacteria of the thiobacillus and ferrobacillus ferrooxidan types, found in the groundwater at the site, derived their energy from oxidizing pyrite or ferrous sulfate.

It was hypothesized that these bacteria catalyzed the oxidation of pyrite in shale, producing sulfuric acid. This sulfuric acid slowly dissolved the calcite in the rock, altering it to gypsum. The gypsum migrated in a much larger volume and exerted pressure on the structure above.

At the St. Luke Church in the New Edinburgh area of Ottawa, Ontario, extensive heaving was reported (22) in the basement floor, and it was suspected that this was caused by the expansion of pyritic shale. The building is underlain by a black carbonaceous shale that contains about 5 percent organic matter, 8 percent calcite, and 4.25 percent pyrite. The investigation also revealed an interesting secondary consequence of pyritic shale oxidation in the form of concrete deterioration caused by the attack by sulfate solution formed by the oxidation of pyrite in the underlying shale.

The church was constructed in 1913, but after 15 to 20 years the basement floor had to be repaired and was covered with a new layer of concrete at the time, because the original concrete, lying directly on the weathered black shale, was severely deteriorated. The uneven thickness of the new concrete suggested that the original floor had already heaved. The floor was reexamined in 1974 and the upward movement was estimated to be 2.5 in. It was confirmed through investigation that heaving had resulted from the growth of gypsum crystals between the bedding planes in the shale.

The mechanism and causes of heave were similar to those already discussed; however, deterioration of the concrete was also significant. Examination of the old concrete with a binocular microscope indicated that the cement had been largely removed, leaving large voids between the aggregate particles. According to Reading (11), when concrete is subjected to acid attack, it is usual for the soluble phases of cement to be removed, leaving a weak porous material, such as what was observed in this case. Calcite is usually a major constituent of old concrete; however, because of the acid attack, it had largely been removed in this case.

## Concrete Deterioration in Oslo, Norway

For more than 60 years the construction industry in Oslo, Norway, has been plagued with the problems of concrete deterioration and foundation upheaval. The unstable forms of the iron sulfide mineral in the shales in the Oslo area are pyrite and pyrhotite (FeS1.4). The rate of oxidation of pyrhotite is extremely rapid. In fact, in its natural state it can be kept in moist air for only a short while. It

appears that pyrhotite can also accelerate the oxidation of pyrite.

Moum and Rosenqvist (30) report that normal sulfate attack may take several years to cause any noticeable damage, whereas water from the alum shale containing ferrous sulfate has caused considerable deterioration in a few months. For example, the concrete walls of an underground bomb shelter built in an alum shale area softened and deteriorated in about 9 months.

According to the authors, the mechanism of attack is closely related to the composition of sulfate solution entering into the system. In the alum shale the solution contains bivalent iron, which is later partially oxidized to trivalent iron. There are differences in the types of effects produced by sulfuric acid, sodium sulfate, and bivalent iron sulfate as they attack concrete. Sulfuric acid mainly attacks the surface of the concrete and dissolves the cement gel and paste. The sodium sulfate solution, on the other hand, may penetrate the concrete without immediate reactions at the surface, but the quantities of sulfate brought into the concrete may be much higher. In the interior of the concrete the sulfate will react with tricalcium aluminate (C3A) in the cement to form ettringite crystals, which occupy a much greater volume than the  $c_3A$ . In contrast to these two types of attack, bivalent iron sulfate may penetrate the concrete in the same way as the sodium sulfate solutions, but may carry with it even higher concentrations of sulfate. In the interior of the concrete the solution has a tendency to decrease the pH rather than increase it, thus leading to either a typical sulfate attack or an acid attack.

With respect to the deterioration of special concrete, Moum and Rosenqvist (30) found that the alum shale solutions appear to attack air-entrained concrete more quickly than other concretes. However, sulfate-resistant concretes appear to perform well.

#### METHODS OF CONTROLLING THE PROBLEM

What can be done to deal with pyritic shales? Unfortunately, understanding the problem does not necessarily ensure the findings of an appropriate solution. However, a number of corrective measures have been suggested and used with some success. Some of the methods of controlling the problem include (a) avoiding the sulfide-bearing shale, (b) controlling or inhibiting the oxidation of the sulfide minerals, (c) site grading to remove or to deeply bury the sulfide carrier, (d) coating with impervious material such as bitumen, (e) oxidation of the layer before construction, (f) building a structural floor system, (g) rock bolting to pin down the floor, and (h) laying a limestone aggregate base.

Avoiding the sulfide-bearing material would solve the problem, but in many instances this is out of the question. It is necessary, therefore, that a solution be found to counteract or control the heaving problem.

Controlling or inhibiting the oxidation process has proved to be a method that provides a satisfactory prevention of expansion. Shales have been flooded with a potassium hydroxide solution in Ottawa (31). It is reported that such measures have effectively counteracted heaving. This basic solution stops the growth of autotrophic bacteria that catalyze the various stages of alteration of pyrite to gypsum. It was found that 0.01 and 0.1N KOH solutions caused swelling to cease, but 1N KOH actually accelerated the rate of swelling. Along the same lines, Dougherty and Barsotti (12) found that less

rapid swelling took place with a sulfuric acid solution (pH 3.5) than with distilled water (pH 7). The instability of pyrite under basic solutions suggests that the submergence of shales in strong KOH solutions may be counterproductive by raising the pH beyond the stability field of the pyrite. In view of this, it would appear preferable to flood the shales with neutral or even slightly acidic solutions to stop the swelling.

Site grading to remove the problem material or to bury the sulfide carrier is another viable solution. Changes in the grading plan have often been used effectively. At some sites the simplest scheme is to lower the grade to completely remove the expansive material or to raise the grade above the swell-prone material to allow swell to occur, with no damage to the structure (2). Obviously, in many instances this solution may be the simplest and most cost effective. However, in other cases the depth or thickness of the expansive layer may make site grading uneconomical.

Another possible means of controlling the problem has been bitumen coating to prevent oxidation of pyrite. For years contractors and builders in the Cleveland area have followed this procedure successfully where the presence of pyrite is known or suspected (32). As part of this procedure, the shale is cut 4 in. to 1 ft above its final grade, and the entire excavated area, including the walls of the cut, is immediately given a spray or brush coat of bitumen. After all other miscellaneous work is completed and the structure is ready for concrete work, the shale is then cut in sections to the proper subgrade depth. As each section is exposed it is concreted immediately. The principal reason for this procedure is to eliminate contact between the shale and air and water. Experiments by Dougherty and Barsotti (12), as well as actual field findings by Quigley and Vogan (21), suggest that the total submergence of shale in groundwater alone will accomplish the same effects because it effectively cuts off the much needed supply of oxygen to the sulfide mineral.

Deliberate acceleration of the oxidation before development is another possibility. However, it has been noted that it would be difficult, if not impossible, to determine the location and extent of all potentially expansive materials (12). Furthermore, even if accelerated greatly, the oxidation process may take too long, making this method uneconomical in terms of actual building time.

Dougherty and Barsotti (12) have also suggested a structural floor system as a solution in some cases. A space provided between the floor slab and the expansive material would actually allow the material to swell with no adverse side effects. The footings can be placed below the potentially expansive stratum or can be designed for a dead load that is sufficient to resist heaving. As noted earlier, a structural floor system will be constructed to correct the heaving problem in the WVU Engineering Sciences Building.

Finally, two other methods proposed by Dougherty and Barsotti (12) are rock bolts and laying a limestone aggregate base. For lightly loaded structures, rock bolts could be used where bolts would be drilled into sound rock below the stratum containing the potentially active sulfide minerals and would serve as an anchor to resist uplift. In laying a crushed limestone aggregate base placed beneath the floor, the thinking is that the limestone would retard the chemical reaction and formation of melanterite. However, gypsum might form from the downward leaching of calcium and might compound the problem.

#### RESEARCH NEEDS

Although the causes and mechanisms of swelling in expansive pyritic shales have been generally identified, insufficient research has been done on methods of controlling or eliminating the problem. Additional research in this area is warranted. Attention should be directed to how soon the reaction may start and at what rate it proceeds after excavation and construction. Further research should also focus on testing and analytical methods of predicting the problem materials and the extent and rate of expected expansion and associated damage. One area that deserves special attention is the morphology of gypsum crystals (i.e., bundles versus flat, bladeshaped crystals) and possible physical and chemical means of controlling the type of crystal formed.

Biochemical means should be explored as a possible way to control heave. If biogenic oxidation can be halted, this might break the chain of oxidation reactions that cause swelling. Possible ways of achieving that may include chemical disinfection to kill the bacteria or creating an unfavorable environment for bacteria growth.

The sulfate attack problem on concrete from pyritic shales should also be studied further. A related problem warranting research is the environmental impact of the leachate from pyritic shales.

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