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Swelling Potential of Dam Foundations Due to Pyrite Oxidation

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ABSTRACT

The form of iron sulfide most abundantly found in nature is pyrite (FeS_2). Commonly recognized by many as “fool’s gold”, pyrite has been identified in igneous, metamorphic, and sedimentary geologic formations dating from Pre-Cambrian to present-day, but is most frequently encountered in dark, carbonaceous sedimentary rocks. Where left undisturbed and unexposed, pyrite is a relatively stable mineral. However, the performance of construction activities in formations containing pyrite, or using materials excavated from such formations, can trigger reactions which may have severely adverse impacts to structures.

While sulfate attack on cementitious materials is a familiar topic in civil construction and related industries, engineers, contractors, and regulators are often less aware of the potential for foundation deflection due to oxidation of pyrite in foundation materials underlying a structure. Where natural or constructed foundation materials include a befitting combination of minerals and are expected to experience less-than-saturated moisture conditions, structures may be subjected to enough heave force to cause multiple inches of foundation heave. Risk can be further increase by the introduction of differential foundation deflection, as foundation surfaces and resisting weight can vary across a project footprint. Concrete spillway integrity may be compromised by cracking which leads to displaced surfaces and concentrated seepage paths may develop along separated concrete joints or where foundation deflection results in water stop failure.

Designers and contractors can avoid such unfavorable performance by sufficiently characterizing project sites and understanding the mechanisms which generate the subject foundation heave. Subgrade design can be tailored to limit desaturation of susceptible formations and spillway structural configurations can be focused toward targeted distribution of weight on subgrade. This paper describes the forms of pyrite which can be encountered in black shales, briefly explains the pyrite-related chemical processes which produce foundation heave, provides suggestions for investigating project site conditions and for accommodating foundation heave potential in design, and presents a case history where a new RCC embankment overtopping spillway design was configured to limit the potential for heave of the black organic shale on which the spillway stilling basin will be founded.

I. INTRODUCTION

It has been commonly understood in the civil industry for many years that sulfates have a harmful chemical effect on concrete. However, issues related to sulfur-induced foundation heave remained a topic of limited discussion until the 1970s [1] when earlier cases of expansion related to sulfur were documented for example sites in Canada [2], Missouri and Kansas [3], New York [4], and a limited number of other locations. Focus on understanding the cause of damage to structures revealed that foundation materials can swell due to the presence of ferrous (pyritic) sulfide minerals, such as pyrite, pyrrhotite, and marcasite.

While iron sulfide minerals are known to occur in a variety of geologic settings, the environment where pyrite and similar minerals are found in greater abundance tends to be within well-bedded, fine-grained sediments which are darker in color and have relatively higher organic/carbonaceous content [1]. Berner [5] describes the process of pyrite formation as occurring in dark, shallow sediments where oxygen is depleted and bacteria, which are sustained by energy from the constituent organic matter, chemically reduce interstitial sulphate to produce hydrogen sulfide (H_2S) that reacts with residual iron in the sediment to form iron monosulfides, such as pyrrhotite, which typically transform to pyrite readily during early diagenesis of the sediments. While this is a simplified and imprecise explanation for a complex process, which is more expansively explained by Sawlowicz [6], readers are encouraged to note foremost that pyrite and other iron sulfide minerals may be encountered on a relatively higher frequency in black shale formations which formed from organic-rich sediments deposited in shallow marine and estuarine environments with sufficient amounts of available ferrous and sulphate minerals and bacteria to reduce these minerals [7].

In North America, numerous pyritic shale units formed during the Devonian geologic period (about 415 to 360 million years ago) in relatively shallow marine environments when portions of the continent were submerged under ocean waters. The approximate locations of Devonian black shales in North America are shown in Figure 1 [7].

Figure 1. Approximate locations of Devonian shale formations in North America [7]

The magnitude of the area in Figure 1 representing the approximate extents of Devonian shale presence suggests that there are a great number of structures, including dams, which may be at risk of experiencing structural damage due to foundation swelling related to the presence of sulfide minerals. It is important to note, however, that the presence of pyrite does not solely create a risk; there are additional factors which impact the potential for volume change in foundation materials.

II.PYRITE OXIDATION AND REACTION WITH CALCITE

As discussed in the previous section, pyrite and other iron sulfide minerals most frequently form in environments where iron and sulfur constituents are submerged. Sulfur inclines to form as sulphate where exposed to oxygen, such that sulfides minerals cannot form in atmospheric conditions. Sulfide minerals are, consequently, encountered in rocks which formed in anoxic environments and may only be preserved in saturated conditions which are bereft of oxygen [8].

Where subjected to a combination of oxygen and moisture, sulfide minerals oxidize to produce ferrous sulfate and sulfuric acid [9]. The oxidation of pyrite and other sulfide minerals is a complex process which involves chemical and biological reactions and varies with site conditions. For simplicity, this paper will refer simply to the oxidation reaction without consideration for chemical or biological impacts to reaction rate. The oxidation reaction alone causes no volume change, but results in the production of [sulfuric] acid runoff which can lead to problems such as contaminated drinking water, disrupted growth and reproduction of aquatic plants and animals, and corrosion of infrastructure composed of metal, concrete, and other susceptible materials. Where calcite is present, it reacts with the sulfuric acid to produce gypsum, which neutralizes the acid and results in a crystalline volume of about twice that of the source calcite and the generation of a significant expansion force during growth of the gypsum crystals [2]. This neutralizing reaction can occur with select clays and calcium salts other than calcite, but the reaction with calcite has been given particular attention [10].

III. MINERAL AVAILABILITY AND MECHANISMS FOR INDUCING SWELLING

Pyrite oxidation and subsequent related reactions are governed by the chemical properties of each respective formation constituent and substances produced in each reaction, but the potential for the occurrence of these chemical exchanges and the reaction intensity depends on the surface area available for reaction and, accordingly, the access to the minerals, the distribution of the mineral in the formation, and the mineral form. The location of the mineral within the formation structure and distribution of the mineral within the formation affect the potential for the mineral to be available for reaction. Minerals which are contained in the interstitial spaces of a lightly fractured black shale formation, such as fine pyrite grains which form between grains of organic-rich sediment, are expected to be less available for reaction than mineral deposits contained exclusively within one or more discontinuities in a formation, such as a pyrite veins which occur along the edges of a moderately-weathered igneous intrusion. However, in the same example, if the black shale formation were highly-fractured and the fine pyrite grains were well-distributed throughout the rock matrix and the contacts along the igneous intrusion were well-preserved, the fine pyrite grains in the black shale may be more readily-accessible than the isolated and protected pyrite veins. Multiple published studies suggest that swell potential can be estimated based solely based on pyritic sulfide content, with some suggesting that as little as 0.1% sulfide content can cause structural damage, but such notions are presented with little to no regard for accessibility of sulfide mineral [11]. A reaction can only occur on the surface of a mineral if the mineral surface is available for contact exposure.

The intensity of pyritic oxidation and quantity of sulfuric acid produced from the reaction is a function of the surface area on which the reaction occurs. Pyrite and similar sulfide minerals occur as larger crystals (“lumps”) and as microcrystals (“seeds”) with diameters between 0.5 and 3 μm. The microcrystalline seeds often agglomerate to form framboids with diameters between 2 and 40 μm [1]. As the total surface area of the seeds contained in a framboid is greater than that of a lump of similar size, oxidation of the framboid is expected to produce a more intense reaction. Examples of these forms are shown in Figures 2 through 4 below.

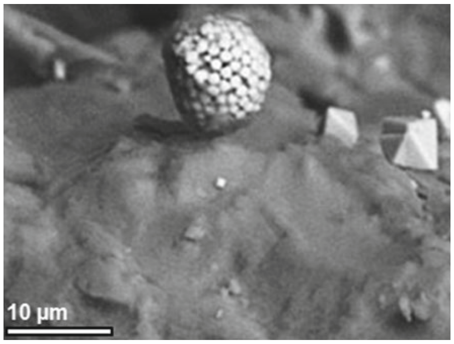


Figure 2. Microcrystalline seed in front of framboid next to octahedral lumps [1]

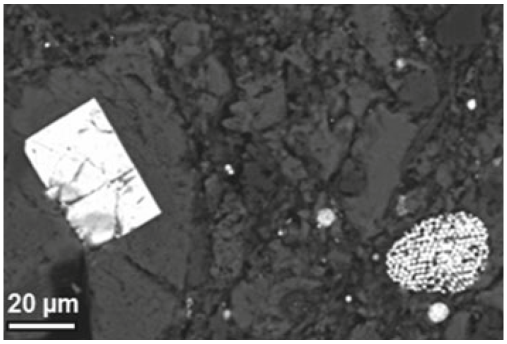


Figure 3. Cubic lump and framboidal pyrite [1]

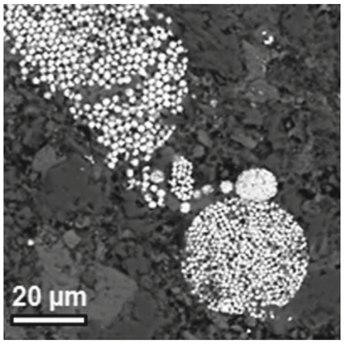


Figure 4. Multiple framboids [1]

As discussed above, the oxidation of pyritic and other similar sulfide minerals is initiated by exposing the mineral surface to oxygen and water. This exposure can occur by introducing oxygenated water to pyritic sulfur in a saturated, anoxic environment, by desaturating the anoxic environment and exposing the pyritic sulfur to atmosphere, or by introducing water to pyritic sulfur in nearly dry conditions. Intermittent exposure can result in a slow oxidation process and development of swelling. One such case of slow oxidation and swelling process occurred at the Beech Fork Water Treatment Plant (Plant) in Clay City, Kentucky. The Plant was constructed in the late 1980s to serve residents in Powell County, Kentucky, which is in an area of the state where the surficial rock formation often consists of the Chattanooga Shale (also called the Ohio Shale and New Albany Shale), a brownish black to black Devonian shale known to contain carbonaceous material and pyrite nodules, in particular abundance in the vicinity of Powell County. Following about 20 years of operation, plant operators began noting heaved flooring and cracked walls in 2010. Following a geologic investigation, it was concluded that groundwater from the adjacent hillside and seepage from the reservoir impounded by a dam adjacent to the Plant gradually introduced moisture to pyritic nodules contained in typically unsaturated weathered shale and disintegrated shale fill on which the Plant was founded [12] and swell of the foundation materials correspondingly increased slowly over time. Figures 5 through 7 show foundation materials uncovered during the investigation, courtesy of Bell Engineering of Lexington, Kentucky.

Figure 5. Weathered rock fragments with yellowish sulfur-stained surfaces

Figure 6. Weathered rock subgrade with visible pyrite nodules

Figure 7. Intact Weathered Rock Fragment with Abundant Iron Sulfide Nodules

IV. INVESTIGATIONS FOR SWELL POTENTIAL

There are a variety of investigative procedures and tests which can be performed to determine the swell potential for subgrade materials. A desktop study of regional published geologic information, such as 7.5-minute geologic quadrangle maps, may reveal indications of trends for mineral content in the vicinity of a site. Following the desktop study, the site should be visited to look for indications of the growth of hydrous sulfate crystals, such as gypsum (calcium sulfate hydrate, $CaCO_3{\bullet}2H_2O$), which indicate the presence of sulfuric acid and calcium carbonate that reacts with the acid to form crystals. For sites with existing structures, a comparison between recent and historical survey data could indicate deflection of a structure since its original construction.

Testing can be performed on rock and soil specimens sampled from foundation materials to determine calcium carbonate and pyritic sulfide content in subgrade materials and to evaluate amount of weather already experienced by constituent minerals. If the depth or elevation to which dewatering will draw down groundwater is known, foundation samples should be obtained from the surface to an elevation below the planned elevation for dewatering so that the designer can understand the variability of conditions in the elevation range where degree of saturation may vary. Soil and rock specimens selected for testing should be sealed in air-tight containers or bags. Care should be taken to avoid wetting specimens, to remove as much air from containers as possible, and to avoid exposing the specimens to hot temperatures. The goal is to preserve the specimens and any constituent sulfur minerals in the sampled condition, which may be altered by exposure to air, water, and hot temperatures. Specimen exposure can be further limited by transporting specimens to the laboratory as quickly as possible.

The currently common method for initially evaluating swell potential is to perform a chemical analysis to determine pyritic sulfide content [11]. This evaluation is accomplished using the ASTM-EPA combined method, as described in the Pennsylvania DER Overburden Sampling and Testing Manual [13], where total sulfur, sulfate, organic sulfur, and calcium carbonate contents are measured and pyritic sulfide content is calculated as the difference between total sulfur and the sum of the sulfate and organic sulfur contents. The measured/calculated values of sulfide and calcium carbonate can aid in rough assessment of swell potential. Simple acid-base stroichiometry can indicate the greater potential for acid rock drainage or expansive hydrous sulfates production. However, accessibility of the minerals must be considered regardless of measured/calculated content and acid-base reactions

Scanning electron microscopy (SEM) and energy dispersive x-ray spectroscopy (EDS) can be used to better understand the forms of pyritic sulfur, the degree to which sulfide minerals have been altered (oxidized), and to more precisely measure the content of various minerals in a specimen. The SEM method produces highly magnified images which allow for observation of individual microcrystalline seeds, framboids, and larger crystals to improve understanding of available surface area and reactivity. SEM images additionally indicate sulfide crystals which have been altered [14]. An example of an SEM image is shown below in Figure 8.

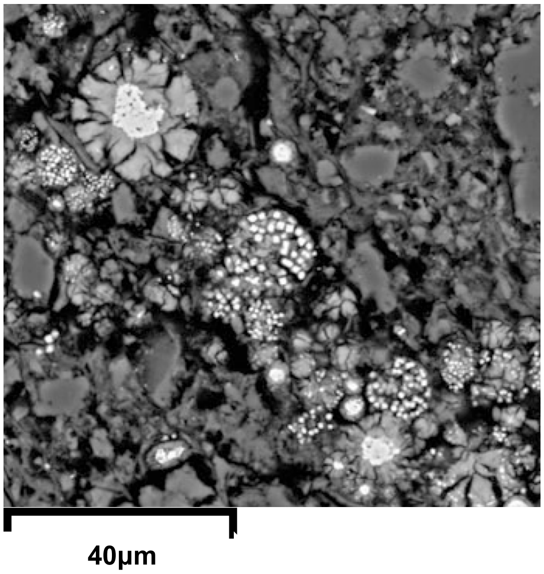


Figure 8. Backscattered electron image showing unaltered (white) and altered (light gray) pyrite framboids [14]

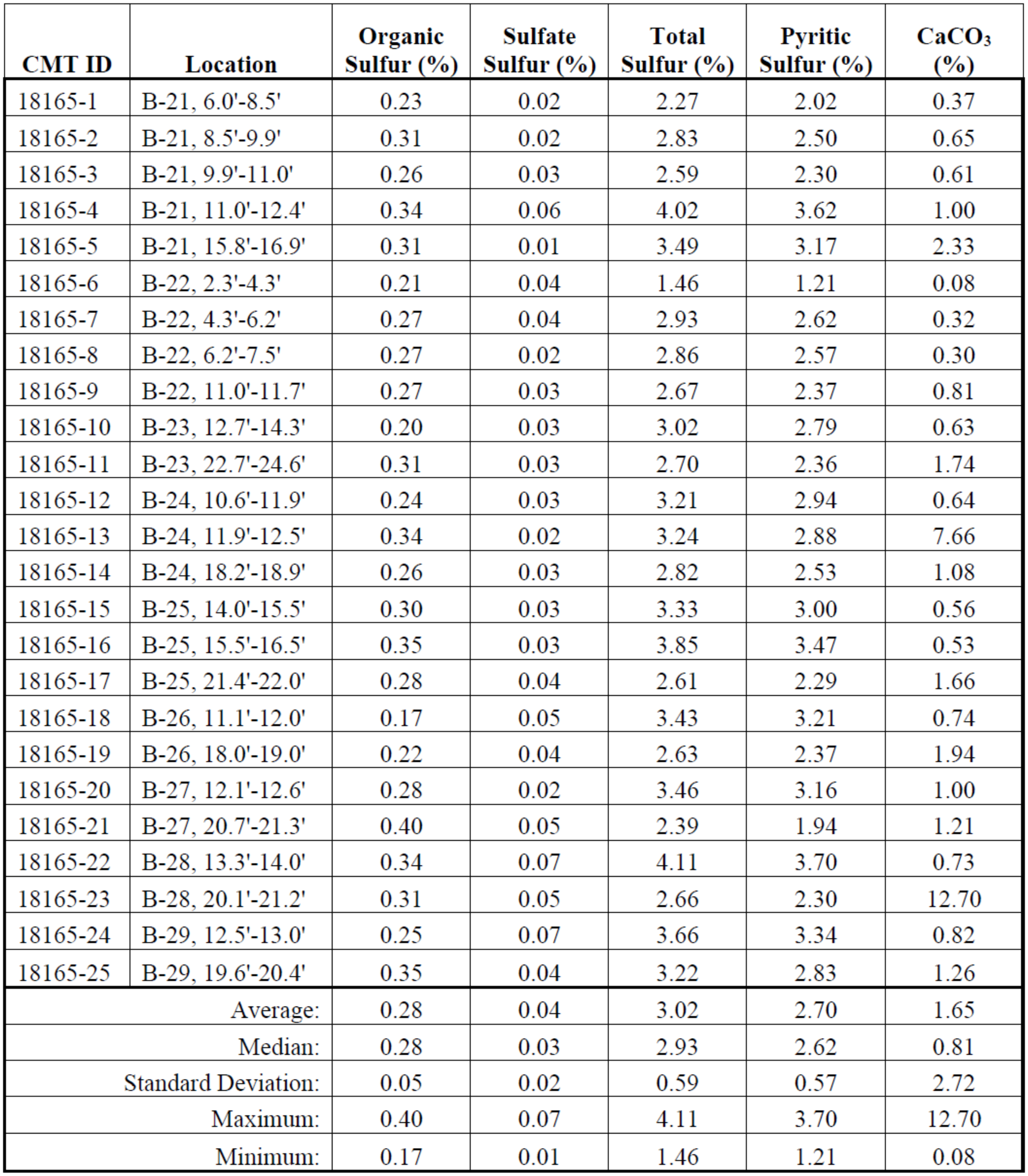
V. DESIGN CASE HISTORY

A dam rehabilitation project in central Kentucky includes the construction of a new roller-compacted concrete (RCC) auxiliary spillway (ASW) over the existing embankment dam. The energy dissipator of the RCC ASW will be founded on the dam's foundation bedrock, which consist of New Albany Devonian black shale. Based on recent structural damage related to foundation swell in nearby municipalities and on the general understanding of the presence of pyrite in the New Albany Shale, the governing agency regulating the rehabilitation design requested that the designer investigate the potential for swell of the rock beneath the dam. Prior to invasive investigations, the designer reviewed published geologic information specific to the vicinity of the site, compared recent and historical survey data for existing structures at the site, and performed a site inspection. Review of survey data indicated that the existing principle spillway reinforced concrete riser and impact basin have experienced little to no movement since original construction of the dam was completed in the mid-1970s. During the site inspection, the designer noted that no signs of sulfate attack were observed on above-ground concrete surfaces and no sulfate crystal were observed on site features, such as drain outlets, or on any natural features, such as stream banks or rock outcrops, in close proximity to the site. Despite indications of limited sulfate activity noted from review of survey data and from the site visit, the designer agreed to perform additional investigations as due diligence.

Multiple borings were performed in the energy dissipator footprint to collect rock core samples for laboratory testing. During construction, the groundwater will be lowered to a depth just below top of rock to facilitate placement of the RCC on a nearly dry rock surface. Accordingly, the borings were advanced to a depth of 15 feet below top of rock to understand the thickness of weathered rock and to evaluate the variability of pyritic sulfide content with depth and within the zone which may be affected by changes to groundwater. A photograph of a pyrite lump in the rock matrix, as shown on the side of a core sample, is shown in Figure 9 below.

Figure 9. Pyrite lump in rock matrix in side of rock core sample

Rock core specimens obtained from the borings were subjected to laboratory testing, including geochemical testing for forms of sulfur, microscopic imaging of thin-cut rock sections, and accelerated rock core expansion testing. Microscopic analyses included petrographic microscopy imaging, scanning electron microscopy (SEM), and energy dispersive x-ray spectroscopy (EDS). The results of the geochemical forms of sulfur testing and the microscopic imaging (Figures 10 through 18) pyritic sulfide appears to be available but the availability of neutralizing minerals, such as calcite, appears to be limited. Accordingly, it is expected that there is a greater likelihood for the site to produce acid runoff than for the rock to swell if constituent pyrite were exposed to oxygen. This conclusion is helpful for qualitative understanding of rock swell potential, but offers limited assistance in developing the rehabilitation design.



CMT ID	Location	Organic Sulfur (%)	Sulfate Sulfur (%)	Total Sulfur (%)	Pyritic Sulfur (%)	$CaCO_3$ (%)
18165-1	B-21, 6.0'-8.5'	0.23	0.02	2.27	2.02	0.37
18165-2	B-21, 8.5'-9.9'	0.31	0.02	2.83	2.50	0.65
18165-3	B-21, 9.9'-11.0'	0.26	0.03	2.59	2.30	0.61
18165-4	B-21, 11.0'-12.4'	0.34	0.06	4.02	3.62	1.00
18165-5	B-21, 15.8'-16.9'	0.31	0.01	3.49	3.17	2.33
18165-6	B-22, 2.3'-4.3'	0.21	0.04	1.46	1.21	0.08
18165-7	B-22, 4.3'-6.2'	0.27	0.04	2.93	2.62	0.32
18165-8	B-22, 6.2'-7.5'	0.27	0.02	2.86	2.57	0.30
18165-9	B-22, 11.0'-11.7'	0.27	0.03	2.67	2.37	0.81
18165-10	B-23, 12.7'-14.3'	0.20	0.03	3.02	2.79	0.63
18165-11	B-23, 22.7'-24.6'	0.31	0.03	2.70	2.36	1.74
18165-12	B-24, 10.6'-11.9'	0.24	0.03	3.21	2.94	0.64
18165-13	B-24, 11.9'-12.5'	0.34	0.02	3.24	2.88	7.66
18165-14	B-24, 18.2'-18.9'	0.26	0.03	2.82	2.53	1.08
18165-15	B-25, 14.0'-15.5'	0.30	0.03	3.33	3.00	0.56
18165-16	B-25, 15.5'-16.5'	0.35	0.03	3.85	3.47	0.53
18165-17	B-25, 21.4'-22.0'	0.28	0.04	2.61	2.29	1.66
18165-18	B-26, 11.1'-12.0'	0.17	0.05	3.43	3.21	0.74
18165-19	B-26, 18.0'-19.0'	0.22	0.04	2.63	2.37	1.94
18165-20	B-27, 12.1'-12.6'	0.28	0.02	3.46	3.16	1.00
18165-21	B-27, 20.7'-21.3'	0.40	0.05	2.39	1.94	1.21
18165-22	B-28, 13.3'-14.0'	0.34	0.07	4.11	3.70	0.73
18165-23	B-28, 20.1'-21.2'	0.31	0.05	2.66	2.30	12.70
18165-24	B-29, 12.5'-13.0'	0.25	0.07	3.66	3.34	0.82
18165-25	B-29, 19.6'-20.4'	0.35	0.04	3.22	2.83	1.26
Average:		0.28	0.04	3.02	2.70	1.65
Median:		0.28	0.03	2.93	2.62	0.81
Standard Deviation:		0.05	0.02	0.59	0.57	2.72
Maximum:		0.40	0.07	4.11	3.70	12.70
Minimum:		0.17	0.01	1.46	1.21	0.08

Figure 10. Image of tabulated results of geochemical testing for forms of sulfur.

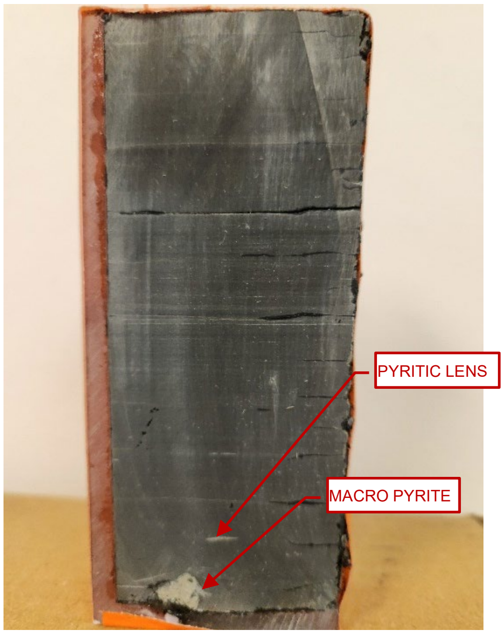


Figure 11. Saw-cut thin section of rock core sample, used for microscopic imaging

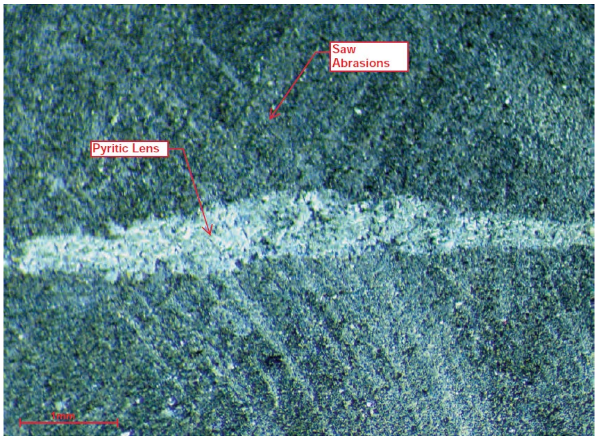


Figure 12. 16X Petrographic microscopy image (scalebar is 1mm) showing a pyrite lens in the rock matrix

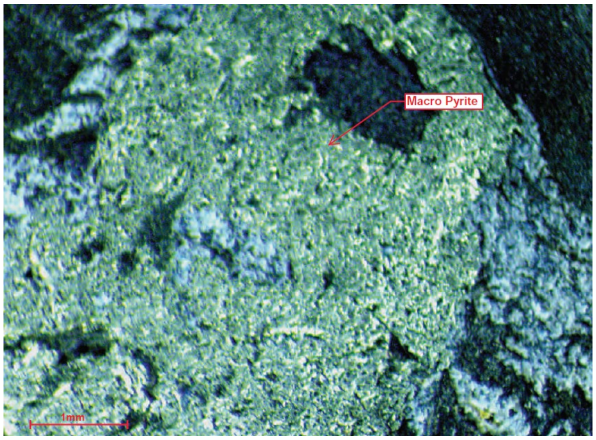


Figure 13. 16X Petrographic microscopy image (scalebar is 1mm) showing a pyrite lump in the rock matrix

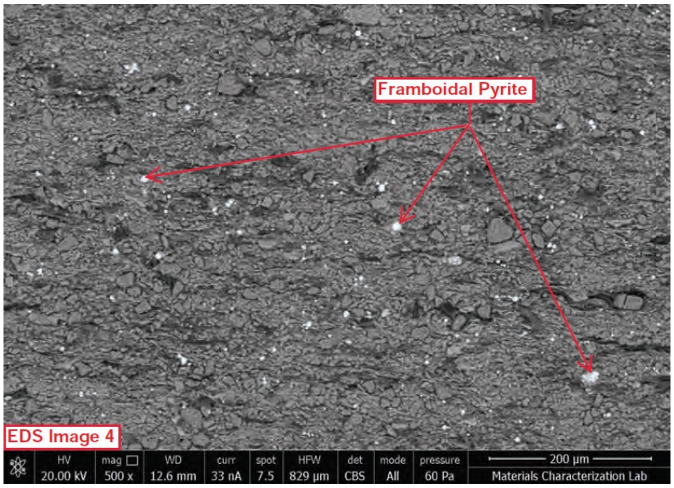


Figure 14. 500X SEM image (scalebar is 200µm) indicating locations of pyrite framboids in rock matrix

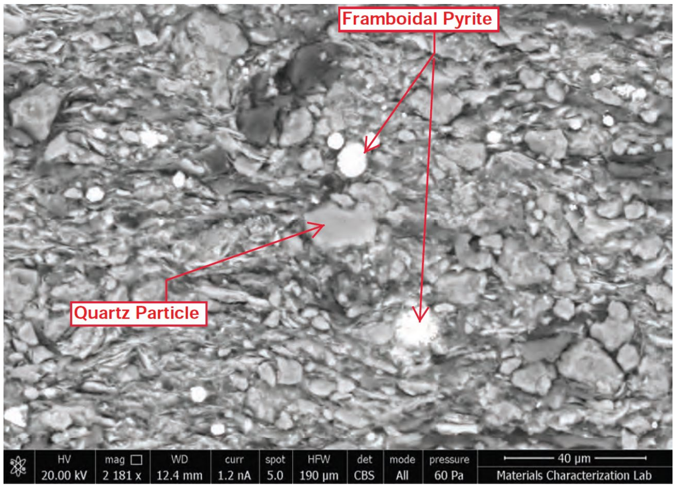


Figure 15. 2,181X SEM image (scalebar is 40µm) indicating locations of pyrite framboids (same sample as Figure 14)

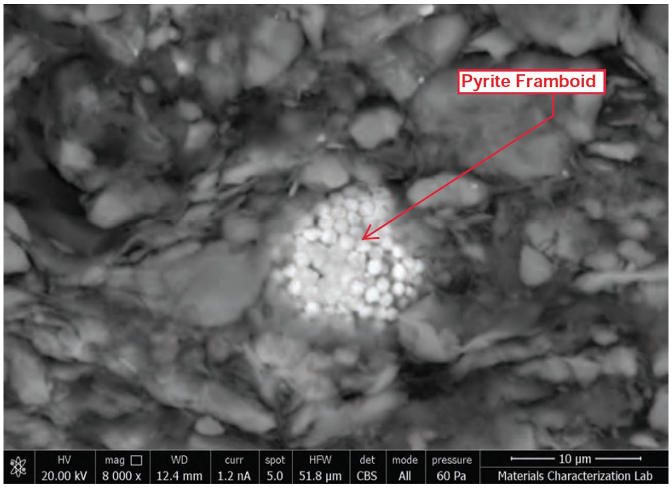


Figure 16. 8,000X SEM image (scalebar is 10µm) of pyrite framboid (same sample as Figure 14)

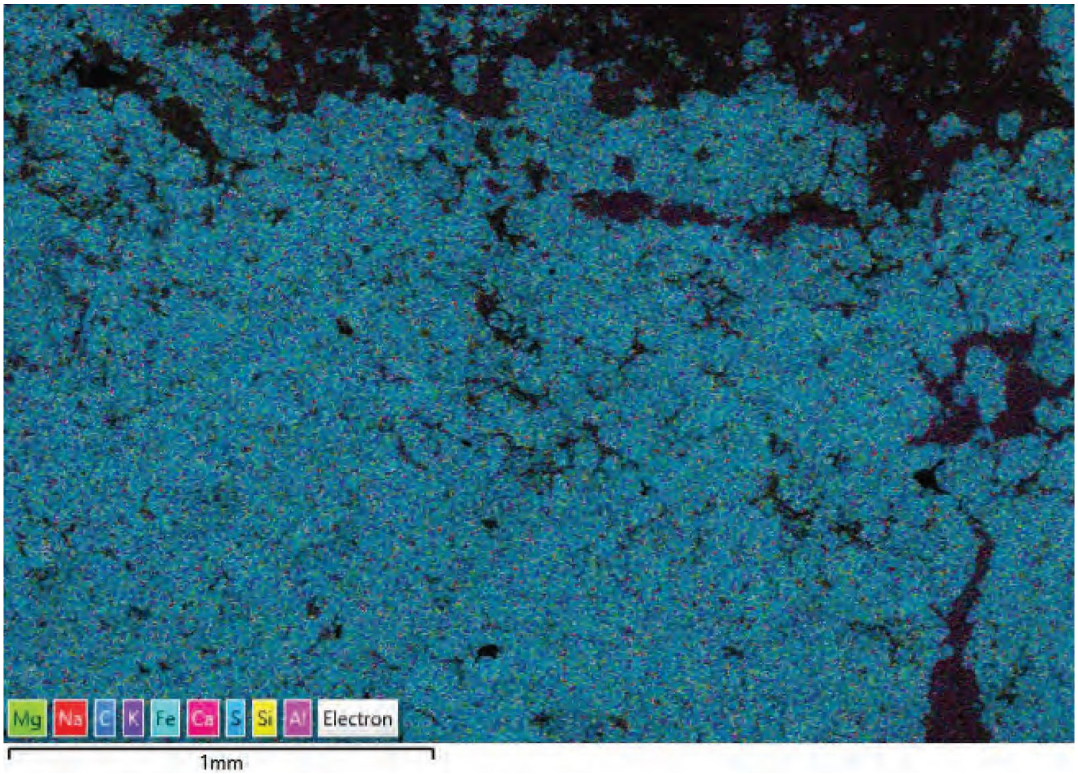


Figure 17. EDS image of material with high concentration of sulfur

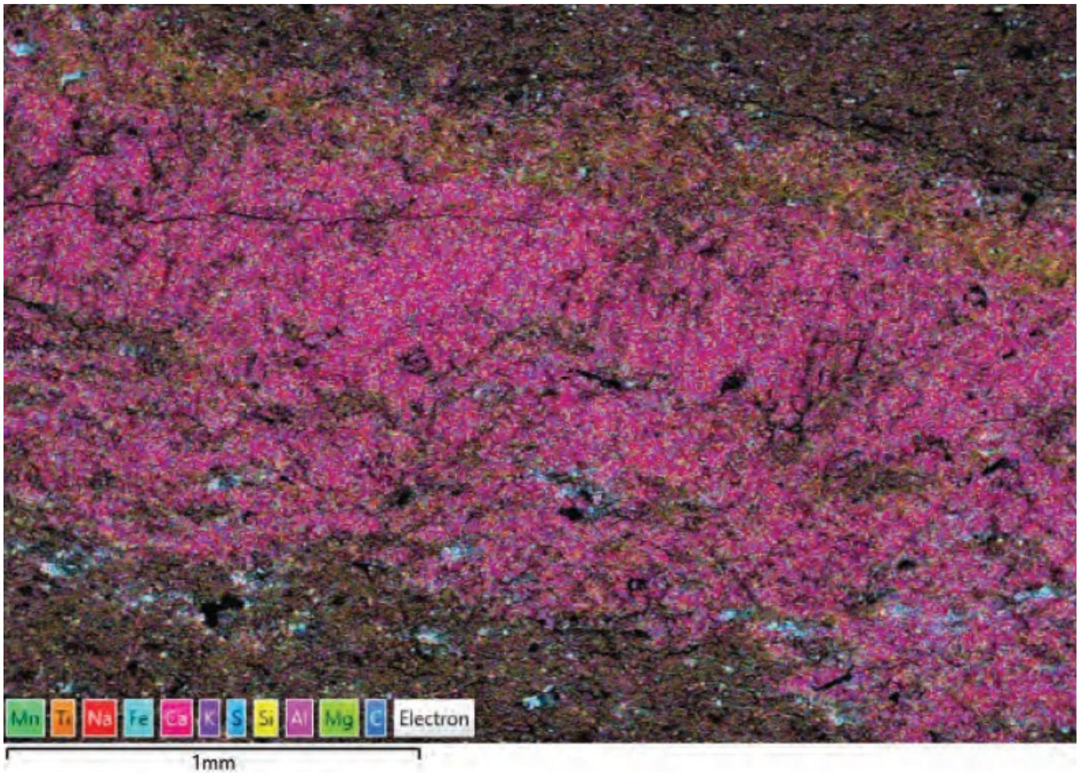


Figure 18. EDS image of material with high concentration of calcium

Accelerated swell tests were performed to derive a relationship between surcharge and strain of rock specimens experiencing swell. The cores were saturated in a 10% hydrogen peroxide (H_2O_2) to accelerate the oxidation of available pyrite and subsequent development of hydrous sulfate crystals which are responsible for expansion forces. During the oxidation and swelling process, a weight was placed on top of the rock core test specimen to simulate surcharge and the vertical deflection due to swelling was measured with a digital gauge with resolution of 0.0005 inches. A photograph of the testing configuration is shown below in Figure 19.

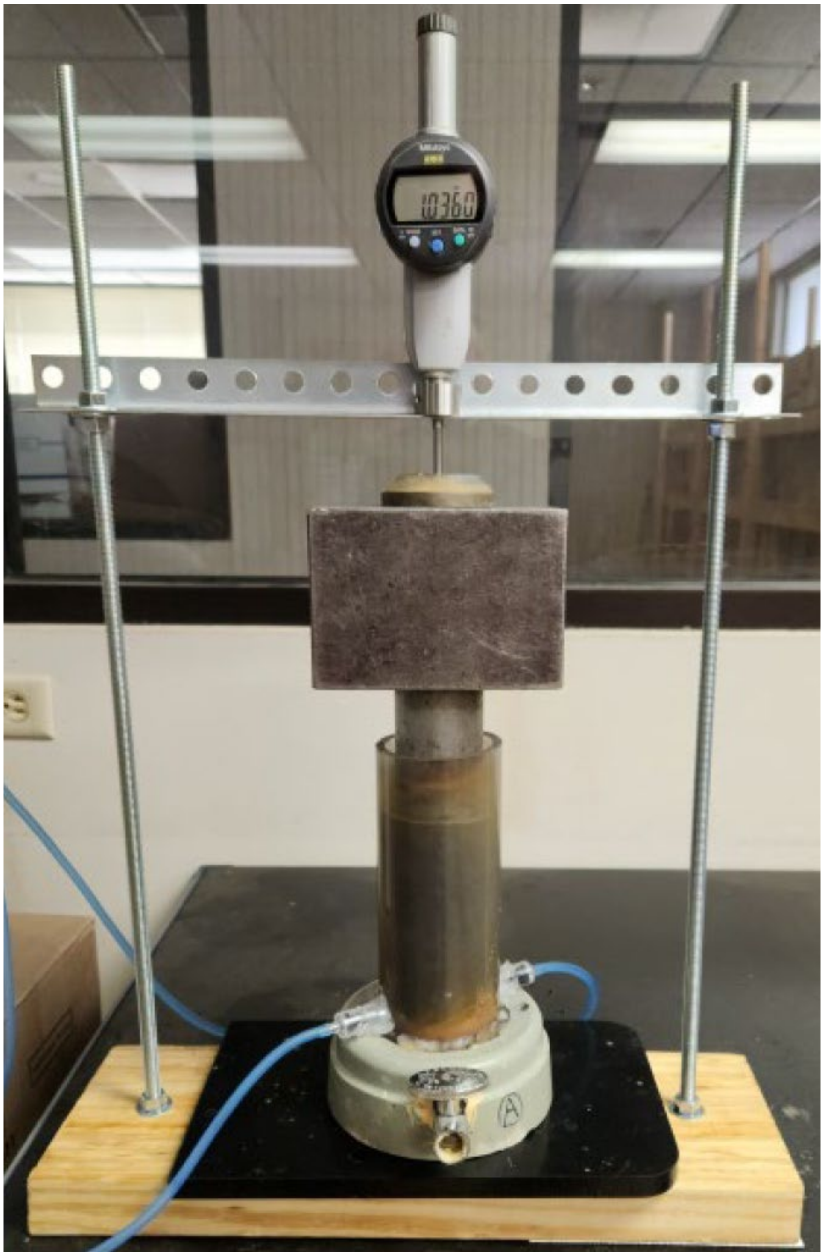


Figure 19. Accelerated swell testing configuration

Seven accelerated swell tests were performed and the measured deflections (ΔH) were normalized by dividing by the initial length (H) of the respective rock core specimens. The results of the tests were plotted with respect to the applied surcharge loads to develop a relationship between normalized vertical deflection and applied surcharge. A graph of the relationship is shown below in Figure 20.

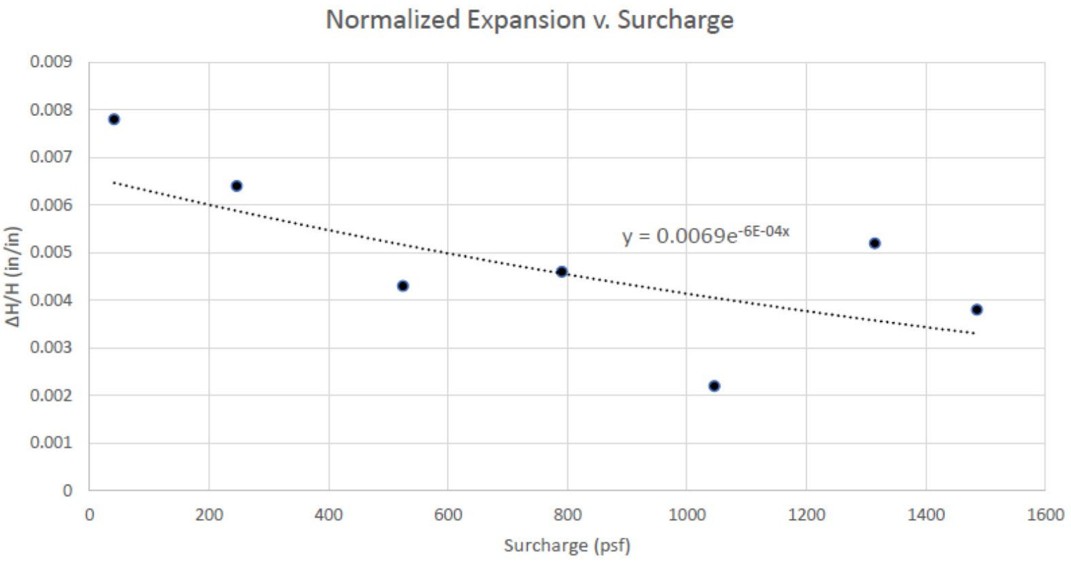


Figure 20. Graph of relationship between normalized vertical deflection and applied surcharge

The results of the accelerated swell testing, specifically the developed relationship graphed above, were used to configure the RCC for the ASW energy dissipator. A thickness of affected rock (H) was conservatively selected based on an assumed thickness of rock which may become unsaturated (i.e. in which atmospheric conditions would be introduced and a minimum required surcharge was iteratively determined using the graphed relationship above with the goal of limiting vertical deflection of the top of foundation rock to one-half inch. A minimum required thickness of RCC was then calculated from the minimum required surcharge. The magnitude of minimum required surcharge for a selected affected rock thickness of 10 feet was determined to be low enough that only two feet of RCC would be necessary for limiting vertical deflection of the top of foundation rock to one-half inch. This design result correlated well with the visual observations noted during the site reconnaissance and with the qualitative conclusions from the geochemical forms of sulfur testing and the microscopic imaging.

VI. CONCLUSIONS/RECOMMENDATIONS

The authors offer the conclusions and recommendations presented below and note that further research is warranted regarding the development of a standard laboratory swell test for pyritic bedrock.

- The potential for oxidation of pyritic minerals depends on the mineral form, access to the mineral, and the potential for introduction of air (for saturated conditions) or water (for near-dry conditions).
- The potential for swell due to oxidation of pyritic minerals additionally depends on the presence of calcite or other minerals which produce an expansive precipitate when exposed to sulfuric acid.
- The use of earthfill which contains pyritic minerals is strongly discouraged, as the mineral surface area is expected to be relatively high and the minerals are expected to be readily accessible.
- Where there is high swell potential of rocks containing pyritic minerals, swelling can be reduced by limiting groundwater drawdown, by avoiding the introduction of air into the rock (e.g. from drilling holes), by limiting the amount of time the surface is exposed, and by applying surcharge load on the rock surface.
- The use of blasting techniques for the removal of pyritic rock can lead to fracturing of the underlying formation thereby increasing exposure of pyrite and other reactive minerals that will lead to greater risk for expansion.
- If pyritic minerals are observed at a project site, the authors recommend that designers engage with a consultant who is knowledgeable and experienced with testing pyritic materials and tailoring designs to account for the presence of pyritic minerals.

VII. REFERENCES

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