

PHOSPHATIC CLAY SLURRIES FOR REDUCING ACID MINE DRAINAGE
FROM RECLAIMED MINE SITES

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Abstract.-- Research at West Virginia University is being conducted on developing a phosphatic clay slurry seal that will prevent or reduce acid mine drainage produced from reclaimed sites. The spoil material that is used to reclaim sites in West Virginia consists primarily of shale and sandstone. When exposed to oxygen and water the spoil produces acid mine drainage. By developing an effective phosphatic clay slurry seal, it will be possible to hydrologically isolate the acid producing materials. An added benefit of this application is that phosphatic clay, which is a waste product of the production of fertilizer, will be utilized, thereby reducing the need for storage of these clay wastes. An effective way of hydrologically isolating the acid producing material is to reclaim sites using a layered system consisting of spoil material, overlain with a phosphatic clay slurry which is in turn overlain by cover soil. Current research is being conducted to define an optimum system that will produce the lowest permeability and reduce the acidity of any effluent generated in the acid materials. To date, laboratory permeability test results indicate that the compactive effort is the dominant variable in determining the permeability of the spoil-slurry system. Low water content slurries (150% to 250%) should be applied to uncompacted spoils, and high water content (250% to 350%) slurries should be applied to compacted spoils. The thickness of the slurry does not appear to significantly alter the permeability of the spoil-slurry system. The addition of the slurry has resulted in a 40 to 80 percent decrease in the permeability of the spoil material. The addition of slurry also dramatically decreases the concentrations of iron, manganese, magnesium, aluminum, and sulfates in the effluent. Slight increases in the pH of the effluent were also demonstrated. Additional testing is underway to establish the optimal conditions for obtaining minimum permeabilities.

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In the past 50 years an ever increasing amount of research has been conducted on the control and remediation of acid mine drainage (AMD) produced from both deep and surface coal mines. Many of these efforts have concentrated on the use of crushed limestone or other base materials to neutralize the AMD. These methods, however, required perpetual reapplication to insure sufficient buffering capacity. Furthermore, the application of base materials could only neutralize the acid, but was unable to stop or prevent the acid producing reaction. The increasing environmental awareness of the public and the increasing cost of remediation has made it desirable for the coal mining industry to seek alternative methods for controlling AMD.

To this end, research is being conducted at West Virginia University on the use of phosphatic clay to control the formation of AMD. Phosphatic clay is the waste product of the production of fertilizer. Approximately 400 million tons of this waste are currently stockpiled in Florida, and this amount is expected to exceed 1 billion tons by the year 2000 (Lin). Using this waste product for the control of AMD will have the added benefit of reducing some of the disposal problems facing the phosphate fertilizer industry.

Phosphatic clay is made up of approximately 25% apatite, 60% smectitic clay minerals, and 15% various other phosphate minerals (Renton). The apatite consists of calcium phosphate ($\text{Ca}_5(\text{PO}_4)_3$), which becomes readily soluble below a pH of 7.0. This dissolution frees the phosphate ion, which has a strong affinity for metal ions. The smectitic clay minerals are composed primarily of montmorillonite. When exposed to water, montmorillonite expands several times its original volume, which results in a very low permeability. These two characteristics of phosphatic clay; the ability to release phosphate ions and low permeability, make it extremely attractive for use in controlling the formation of AMD.

It is generally accepted that the oxidation-hydrolysis of pyrite (FeS_2) contained in spoil material and coal refuse produces AMD. When pyrite is exposed to oxygen and water, it dissociates into ferrous ions (Fe^{+2}) and sulfate ions (SO_4^{-2}). The ferrous ion is further oxidized to the ferric ion (Fe^{+3}). Once the ferric ion has been formed it takes over as the primary oxidizing agent, and these reactions do not stop until the pyrite supply is exhausted (Renton). As these reactions progress metals such as magnesium and aluminum, if present, can also be leached from the spoil. The end result from all of this chemical activity is AMD. The quality of AMD can range from low acidity with high concentrations of sulfates and low concentrations of metals, to AMD high in acidity, metals, and sulfates (Kleinmann).

The application of phosphatic clay to reclaimed mine sites has the ability to not only neutralize, but also completely stop this reaction (Renton). An efficient way to achieve this goal is to place a slurry of phosphatic clay over the potentially acid producing spoil. The montmorillonite provides a first line of defense in the form of a hydraulic seal against infiltrating surface water. Because of

montmorillonite's low permeability, any surface water that is not eventually evaporated will require a great deal of time to pass through the seal. Any water that does manage to pass through the seal will react with any exposed pyrite. The resulting acidic environment will dissolve the calcium phosphate. The phosphate ions will react with the ferric and ferrous ions to form almost insoluble iron phosphates. The major oxidizing agent will be removed, and the AMD producing reaction will be stopped (Renton).

The objective of this research is to determine those variables most important in producing phosphatic clay slurry seals that possess low permeability. The information gained from these experiments will be used to develop field scale tests, which will in turn, be used to make recommendations for coal operators to apply phosphatic clay seals to toxic materials, thus reducing AMD.

MATERIALS AND METHODS

Samples of phosphatic clay were obtained from a phosphate mine located near Mulberry, Florida. The phosphatic clay was shipped in watertight 55-gallon containers. Samples of spoil material were obtained from an active surface mine located in central West Virginia. Both of the materials are indicative of the general physical and chemical nature of materials located in the vicinity from which they were obtained. A cover soil was also obtained from the same reclaimed area as the spoil. This cover soil will be applied over the phosphatic clay seal to prevent the subsequent drying and cracking of the seal that will occur if the seal is exposed to the atmosphere. The cover soil will also provide a vertical overburden stress or weight on the underlying seal and spoil, and it will also provide a base upon which to grow vegetation.

Several standard tests were performed on each separate material to determine its physical characteristics. All tests were conducted in accordance with ASTM standards (ASTM, 1985) and consisted of : hygroscopic water content (D2216-80), specific gravity (D 854-83), standard proctor compaction (D 698-78), Atterberg limits (D4318-84), grain size analysis (D 422-63), and hydrometer analysis (D422-63). These tests allowed extrapolation of the results to similar materials not included in this research. Results from these tests are shown in Table 1.

The next step in the research involved developing mixing procedures for the preparation of slurries. Slurries ranging in water content from 150% to 375% by weight were prepared from the phosphatic clay. A prerequisite for acceptance of a slurry at a certain water content for further testing was that it had to have the ability to be poured so that it could be applied in the field.

Before the clay was mixed with water, it was passed through a funnel with a half-inch diameter opening. Clay clods larger than this opening were broken by hand. The clay was then hand mixed with a spatula to negate any gradation that may have taken place. This procedure was followed to

insure reproducible results, and to provide clay clods that could be placed in suspension by standard laboratory equipment. Initially a slow-action two-speed mixer was used to prepare the slurries. The clay however, tended to clump into large clods, and was very plastic. A rapid-action mixer, run at high speed was then used, and this produced smooth slurries. This mixer was used throughout the remainder of the research to prepare the slurries.

The major portion of the research consisted of permeating various configurations of spoil and slurry. A 10.5 cm diameter by 11.5 cm high rigid-wall double-ring permeameter (Figure 1) was used for all of the tests. The bottom of this permeameter consists of two concentric porous stones separated by a steel ring. The diameter of the ring is such that the area of the inner and outer porous stones are equal, so that under ideal conditions, the outflow is divided equally between the two porous stones. The inner ring is isolated from the sides of the permeameter, and therefore, side wall leakage, which produces inflated values for permeability, does not affect this inner stone. All values for permeability were calculated using the inflow to the permeameter. Outflows from the inner and outer stones were also recorded to make sure no side wall leakage occurred. Side wall leakage was indicated whenever the outflow from the outer stone was much greater than the outflow from the inner stone.

Distilled deionized water was used as the permeant liquid, and this was stored in a nonreactive acrylic reservoir. Pressure was supplied to the reservoir by a pressure board, which forced the water out of the reservoir into the permeameter and thereby set up a hydraulic gradient through the vertical axis of the specimen. The hydraulic gradients used in the test ranged from 10 to 200 (cm/cm). Gradients for each test were determined solely on their ability to expedite test results.

Permeability tests were run separately on the spoil and slurry to provide a base upon which to later evaluate the effect that various combinations of spoil and slurry had on permeability. At least two permeability tests were run on specimens of each material to make sure that results were reproducible. Spoil material specimens were permeated in both the uncompacted and compacted states, and slurries ranging in water content from 150% to 375% by weight were also tested.

With the completion of these "base" permeability tests, experiments were conducted to test the permeability of layered spoil-slurry arrangements. These tests were done to determine the effects of slurry water content, slurry thickness, and degree of spoil compaction on the permeability of the layered systems. Tests were conducted using: 1) loose spoil (no compactive effort), 2) compacted spoil (100% standard Proctor), 3) high water content slurries (>275%), 4) low water content slurries (<275%), 5) thick slurries (>3cm), and 6) thin slurries (<3cm). Eight permeability tests were required to cover all possible arrangements of these variables. See Table 2 for the testing setup. Each layering arrangement was

permeated for at least four pore volumes of flow. One pore volume is equal to the volume of voids in the sample. This insured complete saturation of the voids in the sample, and provided a nondimensionalized means of comparing results from different specimens. Upon completion of the test each specimen was extruded from the permeameter. Settlement of the specimen was measured, and any distinguishing features such as cracking of the slurry and rust deposition from the permeameter were recorded. The specimen was then split along its vertical axis in several places, and the penetration of the slurry into the spoil was measured.

During the permeability tests, effluent from several specimens was collected for chemical analysis. This analysis included determinations of pH, alkalinity-acidity, and concentrations of iron, manganese, aluminum, calcium, magnesium, sulfates, and phosphates. Basic elemental constituents were analyzed using atomic absorption, pH was determined using a digital ionalyzer, sulfates were analyzed using the turbidometric method, and colorimetric analysis was used to determine phosphate concentrations. These analyses were done to determine the quality of effluent produced by the spoil, and to determine what effect the addition of phosphatic clay slurry had on effluent quality.

RESULTS AND CONCLUSIONS

Typical behaviors of uncompacted and compacted spoil specimens both before and after the addition of slurry are shown in Figure 2. All of the uncompacted specimens demonstrated decreasing permeability before the addition of slurry because they were found to have settled. This impeded the flow of water, which resulted in the decreasing permeabilities. The compacted specimens exhibited steady to slightly increasing permeability before the addition of slurry because these specimens had swelled. This enabled the water to move through larger flow paths, which resulted in the increasing permeabilities. Before the addition of slurry every specimen exhibited erratic shifts in permeability between individual permeability readings. After the slurry was added an immediate decrease in permeability occurred, and the variation in permeability between individual readings stabilized somewhat. After the initial large decrease, the permeability of the uncompacted specimens tended to decrease, whereas the permeability of the compacted specimens remained the same or increased slightly.

Results from the "base" permeability tests and the eight spoil-slurry permeability tests are shown in Table 2. Before the addition of slurry, the average permeability of the uncompacted spoil was approximately 1.8×10^{-4} cm/s, and that of the compacted spoil was approximately 1.2×10^{-5} cm/s. Tests 2(1) through 2(4), performed exclusively on slurry, show it to possess an average permeability of approximately 3.0×10^{-6} cm/s.

Based on this last result it was expected that spoil-slurry arrangements would achieve permeabilities similar to 3.0×10^{-6} cm/s. Tests 3(1) through 6(2) show that this is not true. Permeabilities for these tests ranged from a high

Table 1. Index Properties for Spoil Material and Phosphatic Clay

Soil	Natural Water Content	Plastic Limit	Liquid Limit	Specific Gravity	Maximum Dry Unit Weight _s (lb/ft ³)	Optimum Water Content
Spoil	12.5	24	31	2.27	105	13
Clay	31.5	35	135	2.80	75	12

Table 2. Soil arrangement and permeability results for "base" and spoil-slurry permeability tests.

Test No.	Arrangement	Slurry Water Content (%)	Slurry Thickness (cm)	Spoil Avg. K (cm/s)	Spoil and Slurry Avg. K (cm/s)	Change in K (%)
1(1)	Loose Spoil	NA	NA	1.67×10^{-4}	NA	NA
1(2)	Loose Spoil	NA	NA	1.4×10^{-4}	NA	NA
2(1)	Slurry	179(L)	5.56(Th)	$*3.63 \times 10^{-7}$	NA	NA
2(2)	Slurry	194(L)	7.18(Th)	$*1.83 \times 10^{-6}$	NA	NA
2(3)	Slurry	329(H)	2.61(T)	$*4.61 \times 10^{-6}$	NA	NA
2(4)	Slurry	389(H)	4.6(Th)	$*5.24 \times 10^{-6}$	NA	NA
3(1)	Compacted Spoil & Slurry	274(L)	3.10(Th)	2.09×10^{-5}	6.12×10^{-6}	70.7
3(2)	Compacted Spoil & Slurry	353(H)	3.71(Th)	2.20×10^{-5}	5.95×10^{-6}	73.0
4(1)	Loose Spoil & Slurry	174(L)	3.43(Th)	2.69×10^{-4}	2.66×10^{-5}	90.1
4(2)	Loose Spoil & Slurry	225(L)	3.93(Th)	1.53×10^{-4}	6.93×10^{-5}	54.7
5(1)	Compacted Spoil & Slurry	187(L)	1.77(T)	3.57×10^{-6}	3.93×10^{-6}	-10.1
5(2)	Compacted Spoil & Slurry	300(H)	2.54(T)	4.08×10^{-6}	3.30×10^{-6}	19.1
6(1)	Loose Spoil & Slurry	187(L)	1.27(T)	2.69×10^{-4}	9.50×10^{-6}	96.5
6(2)	Loose Spoil & Slurry	300(H)	2.11(T)	1.53×10^{-4}	2.77×10^{-5}	81.9

(H) = high water content

(L) = low water content

(Th) = thick slurry

(T) = thin slurry

K = permeability

*For tests 2(1) through 2(4) the permeability listed in column five is for the slurry.

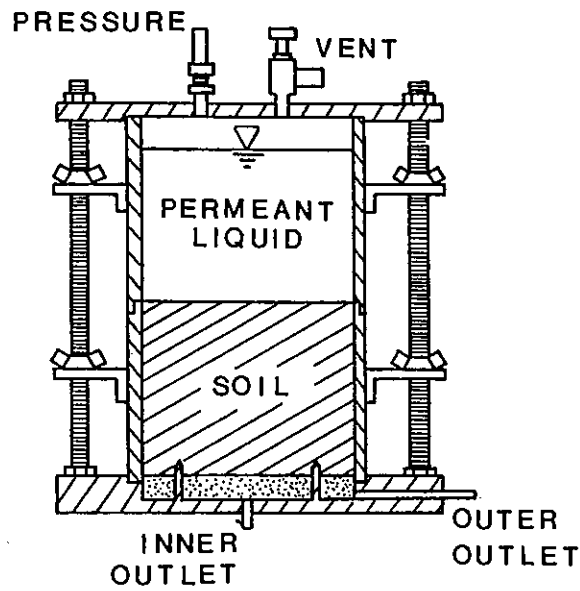


Figure 1 - Double Ring Permeameter

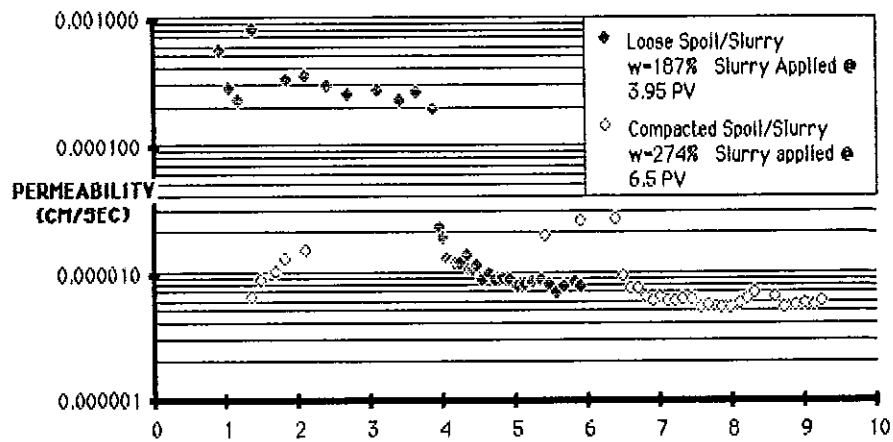


Figure 2- Typical Results From the Permeability Tests on Spoil Material and Phosphatic Slurry

of 7×10^{-5} cm/s to a low of 3×10^{-6} cm/s, with an average of 2×10^{-5} cm/s. All of these tests have average permeabilities higher than that of the slurry by itself. It appears from this result that the spoil plays an important part in determining the overall permeability of spoil-slurry systems. The reason for this result is not readily discernable. Variables used in tests 2(1) through 2(4), such as hydraulic gradient and slurry water content, were similar to the range of values used in tests 3(1) through 6(2). The thicknesses of the slurries used in tests 2(1) through 2(4) were approximately 2 times thicker than slurries used in tests 3(1) through 6(2). This difference however, still does not explain why the permeability of the slurry does not govern the overall permeability because test 2(3), with a thickness of 2.61 cm, has a permeability lower than six out of the eight spoil-slurry tests, which had similar slurry thicknesses.

The results in Table 2, with the exception of test 5(1), show that the addition of slurry will decrease the permeability of the spoil. The magnitude of this decrease, shown in the column labelled percent change in permeability, is not constant. On the average, a 38% decrease in permeability, with the addition of slurry, can be expected for compacted spoil, and an 81% decrease in permeability, with the addition of slurry, can be expected for loose spoil.

Table 3 lists the eight spoil-slurry specimens and their respective final permeability, percent change in permeability with the addition of slurry, and overall performance. Each test was ranked from one to eight with respect to the final permeability, with one representing the lowest permeability, and eight representing the highest. The tests were also ranked from one to eight with respect to the change in permeability, with one representing the largest change, and eight representing the smallest. The percent change in permeability was calculated by dividing the difference between the average permeability of the spoil alone and the average permeability for the spoil-slurry system by the average permeability of the spoil alone.

Table 3 demonstrates that the specimens with the lowest final permeability also tended to exhibit the smallest change in permeability when the slurry was added. Because compactive efforts in the field are often suspect, and uncompacted or undercompacted spoil may be the existing condition, it was equally important to develop spoil-slurry systems that produced low final permeabilities, and large changes in permeability with the addition of slurry. Table 3 allows for the determination of the best possible methods for applying spoil-slurry systems. By adding the point values from the final permeability column and the change in permeability column it is possible to rank the overall performance of different combinations of spoil and slurry. Lower values of overall performance represent better systems.

From Table 3 it can also be seen that the lowest permeabilities were achieved when the spoil was compacted. The reason for this is that compaction reduces void spaces, and flow paths

become smaller and more tortuous. Tests on uncompacted spoil had higher permeabilities because flow paths were not as constricted, and were straighter. As was stated previously, the permeability of the overall system is dependent on both the spoil and the slurry, and not the slurry alone. The addition of slurry can only do so much to lower the permeability of the system. The remaining decrease in permeability comes from the compactive effort applied to the spoil, with increasing effort producing lower permeability.

For the compacted specimens the water content of the slurry did not seem to have as much of an effect on the permeability as did the thickness of the slurry. This result may be misleading however, because the large range in permeabilities between the compacted specimens before the addition of slurry makes it difficult to judge the effect of slurry water content and thickness on the permeability of the compacted spoil. The overall performance ranking of the compacted specimens provided a less biased means of evaluating the effect of these variables on the permeability. Using this ranking, it appears that, in fact, the water content of the slurry was more important than the slurry thickness in determining the final permeability of the compacted spoil-slurry specimens. Higher water content slurries, whether thick or thin, produced better overall performances in the compacted specimens than did lower water content slurries. The reason for this may be that the higher water content slurries can penetrate the compacted spoil and totally or partially clog the flow paths, thereby further hindering water movement. The lower water content slurries may not have been fluid enough to penetrate the compacted surface. Compacted specimens split after permeability testing showed no visible bulk penetration for either low or high water content slurries. The clogging of these flow spaces occurred on the microscopic level however, so blocked flow paths may not have been discernable to the naked eye.

The range in permeability between the uncompacted specimens before the addition of slurry was small, and therefore, correlation between final permeability, change in permeability, and overall performance was much better. As was the case with compacted specimens, the water content of the slurry appeared to have a greater effect on the permeability than did thickness of the slurry. In this case though, lower water content slurries produced better overall performances in the uncompacted spoil specimens. Because the uncompacted spoil had large flow paths, the higher water content slurries may have been flushed out of the specimens instead of clogging the flow paths. Lower water content slurries were less fluid, and may have provided a more stable barrier against the flow of water.

A final conclusion that can be drawn from Table 3 is that the best overall performance was produced from an uncompacted spoil specimen overlain with a low water content slurry. Attempts at compaction should still be made because compacted specimens demonstrated the lowest permeabilities, however, it is still possible to achieve satisfactory results using uncompacted

spoil as long as the slurry covering the spoil has a low water content.

Results from the chemical analysis of effluent are shown in Table 4. Analyses before and after the addition of slurry were only performed on test specimens 3(1) and 3(2). The pH of the effluent for both specimens increased slightly with the addition of slurry, however, it did not reach neutrality. The reason for this result is that phosphatic clay is a relatively neutral material (pH = 7.2) so that it is difficult for it to completely buffer an acidic material.

The affinity of phosphate ions for metal ions is dramatically demonstrated in the large reductions in the concentrations of iron, manganese, magnesium, and aluminum, with the addition of slurry. The absence of any detectable concentration of phosphate ions after the addition of slurry shows that the phosphate ions were completely consumed in various chemical reactions with the metal ions to produce insoluble metal phosphates. A visual inspection of the color of the effluent also indicated that the phosphate ions were effective in removing the metals. The yellow color of the effluent, common in AMD, was apparent before the addition of slurry, however, with the addition of slurry the effluent became clear within one pore volume of flow.

The increase in the concentration of calcium with the addition of slurry is due to the dissolution of the calcium phosphate contained in the phosphatic clay. This result leads to an increase in the hardness of the effluent, however, this consequence must be weighed against the advantages of the large removal of heavy metals.

Sulfate concentrations were also reduced, but the final concentrations were still high. The reductions in sulfate concentration of the two specimens are reflected in the increase of pH for both specimens.

A discrepancy in the iron concentration existed for test specimen 3(2). The initial concentration was very low, but increased significantly with the addition of slurry. An explanation for the initially low concentration may be that the sample of spoil used to prepare the specimen did not contain a representative amount of pyrite. The increase in iron concentration after the addition of slurry may be due to outside sources of iron that were introduced to the specimen during testing. An inspection of the permeameter after the permeability test was completed indicated that spalling of the interior wall had occurred. This could have provided a large amount of iron that flowed down the interface between the permeameter and the specimen.

In conclusion, it appears that the compactive effort applied to the spoil is the dominant variable in determining the permeability of spoil-slurry systems. Larger compactive efforts result in lower permeabilities. Slurry water contents are also important in determining the permeability of spoil-slurry systems. Low water content slurries ranging from 150% to 250% by weight should be applied to uncompacted spoils, and high water

content slurries ranging from 250% to 350% by weight should be applied to compacted spoils. The thickness of the slurry does not seem to be important in determining the permeability of the spoil-slurry system. It may be advantageous, however, to use thick slurries to better insure that any cracks that may develop in the seal do not pass completely through the seal. Before the addition of slurry, uncompacted spoil material has an average permeability of 1.8×10^{-4} cm/s. After the slurry has been applied the average permeability can be expected to drop to 3.3×10^{-5} cm/s. Before the addition of slurry, compacted spoil material has an average permeability of 1.3×10^{-5} cm/s. After the slurry has been applied the average permeability can be expected to drop to 4.8×10^{-6} cm/s. Spoil material should be compacted before the application of slurry. Relatively low permeabilities can still be produced in uncompacted spoil if the slurry water content is kept low. The addition of slurry also greatly reduces the concentrations of iron, manganese, magnesium, aluminum, and sulfates in the effluent through the formation of insoluble metal phosphates. The concentration of calcium in the effluent will increase due to the dissolution of calcium phosphate in the slurry. All of the various chemical reactions will result in a slight increase in the pH of the effluent with the addition slurry.

Results from these tests will be applied to field tests during the summer and fall of 1988. It is hoped that these test will further enable the authors to define the optimum spoil-slurry system, and allow them to develop procedures for applying this technology to the coal mining industry.

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Table 3. Ranking of spoil-slurry tests for the lowest final permeability, greatest change in permeability, and best overall performance, based on results from Table 1

Test No.	Soil Arrangement	Final Permeability(1)	Change in Permeability(2)	Sum of Columns 3 and 4	Overall Performance (4)
3(1)	Compacted Spoil & Slurry	4	5	9	4
3(2)	Compacted Spoil & Slurry	3	4	7	2
4(1)	Loose Spoil & Slurry	6	2	8	3
4(2)	Loose Spoil & Slurry	8	6	14	6
5(1)	Compacted Spoil & Slurry	2	8	10	5
5(2)	Compacted Spoil & Slurry	1	7	8	3
6(1)	Loose Spoil & Slurry	5	1	6	1
6(2)	Loose Spoil & Slurry	7	3	10	5

- (1) 1 = lowest final permeability 8 = highest final permeability
(2) 1 = largest change 8 = smallest change
(3) Numbers determined by adding the final permeability ranking to the change in permeability ranking
(4) Overall performance determined by ranking numbers in column five from lowest to highest, with lowest being the best.

Table 4. Results of Effluent Analysis Before and After the Addition of Phosphate Clay Slurry

Test		pH	Fe	Mn	Ca	Mg	Al	SO ₄ ⁻²	PO ₄ ⁻³
3(1)	Before Slurry	3.89	81.9	248	249	557	623	9320	0.25
	After Slurry	5.73	5.6	17	658	80	6	2145	--
3(2)	Before Slurry	4.14	13	271	300	586	345	8580	0.25
	After Slurry	4.64	160	107	603	78	13	2040	--

Concentrations are in parts per million

