

ASSESSMENT FOR TREATMENT OPTIONS AT THE YOUNG DONG COAL MINE SITE, SOUTH KOREA ¹

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Abstract: The Young Dong Coal Mine site in northeastern South Korea was closed in the early 1990s and initial reclamation was finished in 1995. Even though the adit was filled with limestone, there is still significant acid rock drainage (ARD) flowing from the site. An assessment that was started in March 2008 revealed that there were three types of water flowing from various sources on the site. ARD still flowed from the adit; which carried an average of 500 mg CaCO₃ / L of mineral acidity primarily in the form of Fe(II) with a flow that reached 2.8 m³ / min in spring runoff. This water is the focus of this paper. The hydrology is complex because there are at least two periods of high surface flow during the year, one in the spring and one during monsoon season. The water issuing from the adit is from a diffuse aquifer where concentrations increase when flow increases. In addition, the terrain is quite steep with few level spots. One other factor in the assessment of treatment is that a limestone quarry is only a few kilometers from the site. All of these factors have led to the conclusion that none of the traditional methods of passive treatment can be applied to this site. Instead, it is proposed to use an upgrade of a diversion well that is called a pulsed passive limestone reactor (PPLR). With maximizing all of the parameters that can be changed on a PPLR, it is anticipated that a one stage PPLR system could generate up to 250 mg CaCO₃ / L and so there would be a set of reactors needed to treat the complete flow..

Additional Key Words: Coal mine ARD, limestone reactors, metals contamination,

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Introduction

The Young Dong Coal Mine site is in Northeastern South Korea close to the city of Samcheok. It ceased operation in the 1980s and remediation was done on the site in the 1990s to stabilize waste piles and route water through the site. Much of this remediation concentrated on stabilizing a site that has considerably steep slopes with an overall elevation gain of approximately 100 meters. An attempt to treat the water was made by adding an extensive amount of limestone in the adit before a bulkhead was placed at the opening. However, the water issuing from the adit is acid rock drainage (ARD) that contains extensive mineral acidity in the form of dissolved iron, primarily as Fe²⁺, and aluminum. Besides steep terrain and high mineral acidity, treatment options are limited by the fact that the flow from the adit sometimes exceeds 2.0 cubic meters per min (m³/min).

Table 1. Comparison of Korea water quality criteria (in mg/L) with those of the USA as related to mine drainage.

Water Quality Criteria in mg/L						
Element	Korea			USA		
	Drinking^a	Stream^b	Discharge^c	Aquatic^d	EPA toxic^e	
Ag	-	-	-	0.00015	5.0	
Al	0.2	-	-	0.10	-	
As	0.05	0.05	0.1	0.05	5.0	
Be	-	-	-	0.60	-	
Cd	0.005	0.01	0.02	0.005	1.0	
Cr	0.05	0.05	0.1	0.125	5.0	
Cu	1.0	-	0.5	0.01	-	
Fe	0.3	-	2.0	1.0	-	
Mn	0.3	-	2.0	1.0	-	
Ni	-	-	-	0.2	-	
Pb	0.05	0.1	0.2	0.05	5.0	
Se	0.01	-	-	0.05	1.0	
Zn	1.0	-	1.0	0.1	-	

^a drinking water quality criteria from Korea Drinking Water Management Act

^b stream water quality criteria for human health from Korea Environmental Policy Act

^c discharge water quality criteria for mine drainage at a clean site from Korea Water Quality Preservation Act

^d US water quality criteria for aquatic life recommended by USEPA

^e Extraction procedure toxic level at Resource Conservation and Recovery Act (RCRA)

In order to evaluate the impact of the Young Dong Coal Mine the measured water quality must be compared to some benchmarks. Water Quality Criteria (WQC) are recommended in order to protect human or ecological health and represent one possible set of benchmarks. There are differences in the levels of target elements, which depend on these two objectives of the WQC. Therefore, prior to discussion of the site-specific results, it is necessary to consider the principal aims of the recommendations. WQC for inorganic elements related to freshwater or mine drainage are shown in Table 1. Korean WQC are structured by the type or usage of water. The main purpose of Korean WQC is to protect human health and WQC for drinking water represent the lowest concentration of target elements in the media. Although this provides a benchmark for comparison, the unlikely use of Young Dong water as a drinking water source makes it more stringent than necessary. Stream Korean WQC is also recommended to protect human health and has a shorter list of inorganic elements. The final Korean WQC, with the highest pollutant discharge permission levels, are for discharge from mine adits located in otherwise clean areas and are based on the Korea Water Quality Preservation Act. For this WQC, discharge water includes any water that comes out of any facilities for water treatment. Table 1 also includes WQC for water that has 100 mgCaCO₃/L hardness established by the USEPA that are designed to protect aquatic life (US EPA, 2004). These levels have been established on the basis of available acute or chronic toxicity data for the pollutants. Thus, meeting these levels should result in a safe and sustainable aquatic environment for biota. Finally, Table 1 includes the USEPA standards for leachates obtained from waste materials as established by the Resource Conservation and Recovery Act (RCRA) (US EPA, 2002). Exceeding these criteria will result in the classification of the waste as hazardous and requiring special disposal procedures.

Any treatment initiated depends strongly on the quality and quantity of water flowing from the adit. Also, water flows from some of the remediated waste piles on the site and from other natural streams. Some of these flows appear to be of good quality and others appear to be only marginally impacted with metal contaminants. Thus, a study of the sources, quality, and flows of water at the Young Dong site was initiated so that the water balance can be established. The reconnaissance sampling episode took place in November 2007. Based on the results of this episode, an extensive sampling event took place on March 26-27, 2008. For this trip, personnel from the Colorado School of Mines (CSM) and the Korean Mine Reclamation Corporation (MIRECO) participated. Subsequently, follow-up sampling trips were made by MIRECO

personnel on April 29, May 29, June 30, and in July. Table 2 gives the concentrations of important chemical parameters and the flows from the mine adit for the 2008 sampling dates (Wildeman, et al. 2008). The objectives of this paper are to use the information found so far in 2008 on the flows and chemistry of the adit water to suggest possible treatment options for this water.

Table 2. Concentrations (mg/L) of elements relative to stream and toxicity standards for adit water ARD at the Young Dong site. The values in red are above drinking water standards. “ND” means not detected.

Drinking & Aquatic Element Levels for Young Dong ARD in mg/L							
Element	Det. Limit	Korean Water Stds.		March	April	May	July
		Aquatic	Drinking				
Flow in m ³ / min		mg/L	mg/L	0.48	2.78	2.45	Not meas.
pH				4.52	5.08	5.68	4.43
Al	0.0173		0.2	21.9	2.10	0.062	27.5
As	0.0610	0.05	0.05	ND	ND	ND	ND
Cd	0.0015	0.01	0.005	0.012	0.002	0.002	0.003
Cr	0.0039	0.05	0.05	ND	0.000	ND	0.001
Cu	0.0015	-	1.0	0.010	ND	ND	0.027
Fe	0.0044	-	0.3	195	286	265	292
Mn	0.0005	-	0.3	4.43	5.03	4.47	5.43
Ni	0.0035	-	-	0.243	0.266	0.221	0.368
Pb	0.0137	0.1	0.05	ND	0.007	ND	0.007
Se	0.0504		0.01	ND	ND	ND	ND
Zn	0.0013		1.0	0.206	0.256	0.203	0.284

Proposed Treatment of the Adit Water

Assumed Variables

To make a treatment assessment on the adit water, a number of scientific and engineering calculations, need to be performed. To accomplish these calculations, standard concentrations and flows have to be established. Using the March values shown in Table 2 as a guideline, the following water quality and quantity parameters for the ARD will be assumed.

- Flow of ARD water; 2.0 m³/min, 2,000 L/min
- Concentration of dissolved iron: 210 mg/L
- Concentration of dissolved aluminum: 20 mg/L
- Concentration of dissolved Fe(III): 5.0 mg/L
- Concentration of dissolved manganese: 5.0 mg/L

- pH 4.5
- Mineral Acidity 500 mg CaCO₃/L
- Elevation gain from ARD outflow to the edge of the property: 100 meters

With respect to these assumptions, on April 29, 2008, the apparent peak spring ARD flow was 2.8 m³/min, and, as seen in Table 2, the iron, aluminum, and manganese concentrations were 286, 2.0, and 5.0 mg/L, respectively.

Reaction Stoichiometries

To remove the iron and aluminum, alkalinity has to be added to the water. This could be in the form of hydrated lime as Ca(OH)₂ or limestone CaCO₃. The relevant alkalinity reactions are



Lime give 2 moles of alkalinity for 1 mole of reactant, and, assuming neutralization does not proceed above a pH of 6, then high Ca limestone also gives 2 moles of alkalinity. If limestone is used, the amount of alkalinity needed per liter is 500 mg CaCO₃/L. If lime is used, the amount of alkalinity per liter needed is

$$\frac{500 \text{ mg CaCO}_3/\text{L} \times 74 \text{ mg Ca(OH)}_2/\text{millimol}}{100 \text{ mg CaCO}_3/\text{millimol}} = 370 \text{ mg Ca(OH)}_2/\text{L}$$

Pure lime, CaO, which is not hydrated, could be used for dissolution. However, in a system that does not have constant supervision, the hydration of the lime to Ca(OH)₂ is inefficient and much of the CaO is lost because the solids are flushed out of the reactor. Also, hydrated lime should be slurried with make up water because direct addition causes many of the particles to become coated leading to inefficiencies. In addition, these calculations assume that the hydrated limes and limestone are pure. Usually, the purity of industrial grades of these reactants is about 95 %.

Using the above values for the amount of alkalinity that is needed, the amount of material needed per day for treatment can be calculated. For calcite the amount is

$$\frac{500 \text{ mg CaCO}_3 / \text{L} \times 2000 \text{ L/min} \times 1440 \text{ minutes} / \text{day}}{1,000,000,000 \text{ mg/metric ton}} = 1.44 \text{ metric tons CaCO}_3 / \text{day}$$

For hydrated lime, the amount is

$$\frac{370 \text{ mg Ca(OH)}_2 / \text{L} \times 2000 \text{ L/min} \times 1440 \text{ minutes} / \text{day}}{1,000,000,000 \text{ mg/metric ton}} = 1.07 \text{ metric tons Ca(OH)}_2 / \text{day}$$

The important point of these calculations is that any treatment using an active neutralizing agent requires an extensive operation because at least 1 metric ton of reactant has to be delivered to the site per day. Furthermore, the efficiency of a hydrated lime plant ranges from 40 to 85 % depending on how well the slurry is produced and the type of mixing that is employed. For the pulsed limestone bed process, when high calcium limestone was used, the efficiency averaged 90 % (Hammarstrom, et al. 2003). In a situation such as this, a storage silo could be constructed and the reactant delivered to the treatment system from the silo. The AquaFix Company¹ in West Virginia specializes in such silos and reactor systems for lime and hydrated lime. Their off-the-shelf technology is probably the least expensive way to make a simple active system to treat the ARD. Such a silo could also be used to store and deliver limestone.

Another factor to consider is cost. Based on a per mole basis, and using 1994 cost information (Hedin et al. 1994), limestone costs \$US 0.69 per kilomole of alkalinity, and hydrated lime costs \$US 2.64 per kilomole of alkalinity. In the case of the Young Dong ARD, limestone would have almost no cost because there is a large limestone quarry less than 3 kilometers away and limestone neutralization can use discarded fine-grained material. Based on economic experience from the Argo Tunnel Treatment Plant in Colorado, the cost of hydrated lime would be approximately \$US 200,000 per year (Mary Scott, 2005). Granted, using the Argo Tunnel Plant may not be a proper economic comparison, nevertheless, because treatment will continue far into the foreseeable future, the cost of neutralizing material is an important consideration.

One problem with using limestone for neutralizing ARD is that it dissolves slowly. This causes two problems. A large system is needed to ensure contact of the water with the limestone for at least a day (Hedin et al., 1994, Skousen and Ziemkiewicz, 1995). The second problem with slow reactivity is that the iron and aluminum oxyhydroxide neutralization products coat the limestone and lower reactivity even further (Hammarstrom et al., 2003). Nevertheless, the least expensive treatment option is some system that uses limestone but does not require continuous monitoring or active delivery of the reactant. Because the cost differential at the Young Dong site between an active lime treatment plant and a semi-passive system that uses limestone is so large, an attempt to use a limestone system must be tried. How this system will work and how it compares with existing systems is explained in the following sections.

¹ Any use of trade, product or firm names is for descriptive purposes only and does not imply endorsement by the U.S. government.

The Proposed Treatment System

Examination of the dissolution kinetics of limestone (Plummer et al. 1978; Sverdrup, 1985; Watten et al., 2004) shows that there are three primary factors that will increase the rate of calcite dissolution:

- Increase the hydrogen ion concentration (lower the pH),
- increase the pressure of CO₂ in the system (this CO₂ can come from the calcite itself),
- increase the relative surface area of the calcite particles.

In an attempt to maximize these factors, Drs. Barnaby Watten and Philip Sibrell designed the pulsed limestone bed (PLB) reactor that enabled small particle sized limestone (0.1 to 1.0 mm) to become a fluidized bed. This occurred in a closed reactor that retained the product CO₂, and this lowered the pH (Hammarstrom et al., 2003; Sibrell et al., 2007). This active treatment system has been able to generate alkalinities above 600 mg CaCO₃ / L in the treatment of ARD.

A diversion well is a system that passively uses limestone for treatment (Arnold, 1991; Skousen and Ziemkiewicz, 1995). It uses a head of water flowing through a well to agitate a bed of crushed limestone to generate better dissolution kinetics. However, this type of system does not insure the best fluidization of the limestone bed. What is needed is a diversion well type of system that maximizes calcite dissolution in the manner that the PLB does. The proposed passive pulsed limestone reactor (PPLR) is an attempt to design a passive reactor that will maximize the dissolution of calcite. Table 3 compares the PPLR system with a PLB and a diversion well.

Figure 1 is a diagram of the proposed PPLR system. The first feature that is needed is a mechanism that will ensure that the bed will fluidize so that fine-grained limestone can be used in the system. This is accomplished by the use of a siphon drain that allows the tank to fill and then, when it reaches a certain level, it releases the water in a flush in much the same way that a common toilet operates. Another feature needed to maximize calcite dissolution is a means of retaining the CO₂ that is produced in the treatment tank. The design of this portion of the proposed system is problematic because if water is retained for too long a time, then the limestone may become coated with ferric hydroxide. Consequently, laboratory testing of a prototype PPLR will be necessary to determine how to achieve that highest possible alkalinity.

Table 3. Comparison of the properties of a diversion well, the proposed passive pulsed limestone reactor treatment, and a pulsed limestone bed.

Properties	Diversion Well	Pulsed Passive Limestone Reactor	Pulsed Limestone Bed
Function	Adds Alkalinity	Adds Alkalinity	Adds Alkalinity
Treatment Mode	Passive	Passive	Active
Main Reactant	Limestone	Limestone	Limestone
Other Possible Reactants	unknown	Steel Slag	unknown
Reactant Particle Size	1.25 – 2.5 cm	Below 1 mm	Below 1 mm
Reactor Bed Movement	Sometimes fluidized, depends on flow	Fluidized, intermittently, depends on flow	Fluidized intermittently, on timer
Alkalinity generated using 0.01 M H ₂ SO ₄ (mg CaCO ₃ /L)	~ 50	~ 100	~300
Resistance to Armoring	Low	Medium	High
Flow control	No	Yes	Yes
Capable of retaining CO ₂	No	Yes	Yes

Competing Passive Treatment Technologies

Besides a diversion well, other passive treatment systems have been developed for use on ARD. The passive systems discussed below are all proven technologies. However, there are problems using any type of passive treatment on ARD:

- high mineral acidity, especially acidity due to aluminum in the water,
- high flows,
- no level land spaces available.

It is readily apparent that all of these problems are present with the Young Dong ARD. The following are the passive treatment possibilities listed from the most likely to least likely to work based on the opinion of the authors.

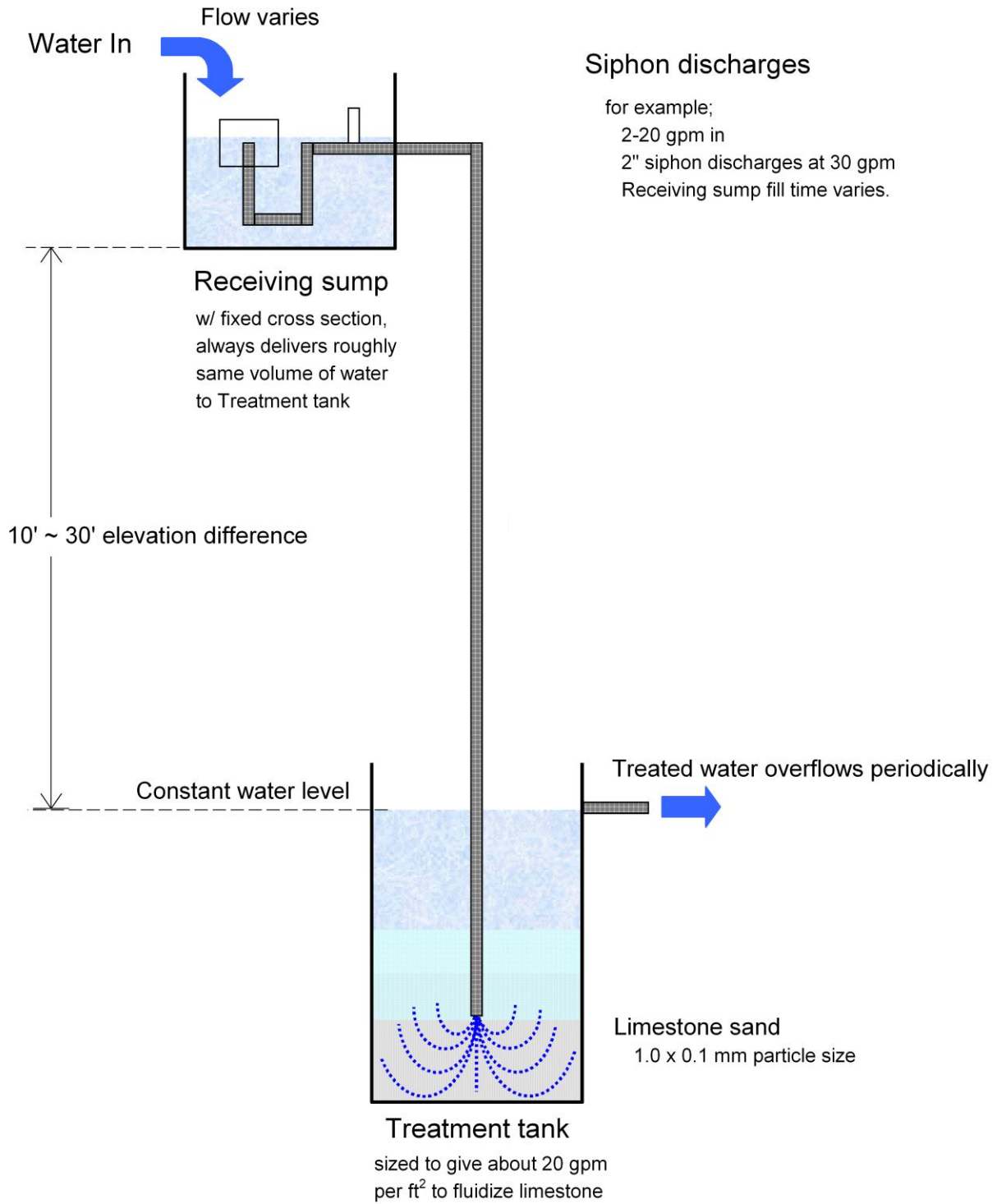


Figure 1. A diagram of a fundamental Passive Pulsed Limestone Reactor

Sulfate Reducing Bioreactor (SRBR)

A sulfate reducing bioreactor uses sulfate reducing bacteria to produce hydrogen sulfide and bicarbonate (Wildeman and Updegraff, 1998). The sulfide will precipitate FeS and the bicarbonate will raise the pH, which will cause the precipitation of aluminum. Such reactors have worked well on waters with high mineral acidity (Gusek et al. 2008). However, the influent flows have been quite low, usually below 80 L/min. In the case of the Young Dong ARD, the area needed for treatment of such a high flow of high mineral acidity water would be large. Using a proprietary sizing program of Golder Associates Inc., to treat the ARD water at Young Dong would take a SRBR of 4.5 hectares in size. One reason why the SRBR is the first choice for passive treatment is because it has not suffered the aluminum plugging problems that the following two systems have experienced. In an SRBR, the aluminum is precipitated as aluminum hydroxyl sulfate, $\text{Al}(\text{OH})\text{SO}_4$, instead of gibbsite, $\text{Al}(\text{OH})_3$ (Thomas, 2002; Thomas et al., 2004, Mitchell and Wildeman, 1996). After water flows through the SRBR, it has to flow through an aerobic polishing cell to oxidize any excess organic material and to reintroduce dissolved oxygen into the water. This will substantially add to the size of the treatment system.

Successive Alkalinity Producing System (SAPS)

A SAPS system is a vertical flow reactor that sends the ARD through an upper layer of organic material to reduce all the iron to Fe(II), and then through a layer of limestone to dissolve it to produce alkalinity (Skousen et al., 1998). After exiting a SAPS system, the water is directed through a settling pond so that Fe^{2+} can be oxidized and precipitated as iron oxyhydroxide. A removal rate of 50 g CaCO_3 acidity / square meter / day is a reasonable average system performance (Rose et al., 2004). Using this value, it would take approximately 30 hectares of SAPS systems to treat the ARD using the quality and quantity parameters listed in Table 1. A big problem with using a SAPS is that they also are prone to clogging with an aluminum oxyhydroxide precipitate (Rose et al., 2004, Hedin et al. 1994). So, with aluminum concentrations in the Young Dong ARD shown in Table 1, if a SAPS system is used, it will probably plug in a matter of years.

Anoxic Limestone Drain (ALD)

An ALD is a passive form of limestone neutralization that uses the same treatment principles as that of the proposed PPLR. A key requirement of an ALD is that all oxygen has to be eliminated from the water before it enters the ALD because if the dissolved iron oxidizes, it will

precipitate and plug the system (Skousen et al., 1998). It is interesting to note that a form of ALD was put into the adit of the Young Dong Coal Mine before the entry was sealed. It is also the case that dissolved aluminum in ARD water will also precipitate in the ALD and clog the drain. Consequently, it is very possible that an ALD at Young Dong will fail. Assuming that it did not plug, the primary engineering parameter for design of an ALD is that the water should be retained in the drain for 24 hours (Hedin et al., 1994). Usually, the submerged drain would be 2 meters in width and 1 meter in height. Using these measurements, a flow of $2.0 \text{ m}^3/\text{min}$, and 50 % porosity in the drain, it would take 3,000 meters of drain to treat the ARD.

Conclusions

Because of the high mineral acidity and considerable aluminum concentration, the Young Dong adit water represents the most chemically difficult type of water to treat passively. In addition, the flow is considerable ($2 \text{ m}^3/\text{min}$) and the terrain offers minimal level spaces. Consequently, most all of the site considerations favor the use of an active lime slurry treatment system. However, because a limestone quarry is only a few kilometers away, use of limestone as the alkalinity source is an extremely attractive possibility. The problem is that the most established passive methods of using limestone (ALD and SAPS systems) would likely be clogged by aluminum precipitates and would need large areas to treat such large flows. Aluminum precipitates would not clog a SRBR, however, the anaerobic treatment area would be at least 4.5 hectares and a comparably sized aerobic polishing system would also be needed. Consequently, a new type of system to use limestone as the reactant is needed.

The PLB process, an active treatment process that uses limestone, has generated over 500 mg/L CaCO_3 alkalinity at other locations. In contrast, the diversion well, an established passive process uses limestone but only generates about 100 mg/L CaCO_3 alkalinity. It is proposed to try to merge the features of these two systems and develop a passive pulsed limestone reactor (PPLR). The PPLR uses a siphon drain to insure a fluidized bed reactor. However, laboratory development of the reactor tank is needed to determine how to retain the produced CO_2 in the system because this is a key factor in developing high alkalinities in the PLB. It is hoped that by maximizing how the PPLR functions that alkalinities of 250 mg/L CaCO_3 alkalinity can be generated and this would make the system viable as a treatment option for the Young Dong adit water. Because the adit water averages 500 mg CaCO_3/L and has a flow of $2 \text{ m}^3/\text{min}$, an array of reactors would be needed to treat the complete flow.

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