# Past, Present and Future for Treating Selenium-Impacted Water (1)

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**Abstract:** Water quality in selected regions of the US is impaired due to selenium, which typically originates from shale formations that were deposited in a marine environment. In the Western US, these shale formations typically underlie agricultural zones or are associated with hard-rock metal mining operations; this results in contaminated runoff and/or seepage. In the Eastern US, the marine shale formations may be associated with coal deposits. Thus, selenium can be a constituent of mining influenced water (MIW) which has been typically referred to as acid mine drainage (AMD). This is somewhat of a misnomer, as selenium is found in some western areas as a contaminant in circum-neutral drainages. Because of the diffuse and widespread nature of this source, often there are only limited opportunities to reduce uncontrolled selenium releases to the environment. One option is to treat selenium-contaminated surface water at strategic locations, such as where point-source discharges occur. Collection and centralized treatment of selenium-contaminated water is a second option based on local site conditions and regulatory considerations.

In many respects, the geochemical behavior of selenium is similar to sulfur; they both share a common group in the periodic table of elements. Early research work in sulfate reducing bioreactors revealed that selenium was reduced biologically in concert with sulfate. At the time, selenium was not a major parameter of concern in coal MIW as the potential selenium impacts were typically masked by elevated acidity.

The paper discusses various past, present and future approaches for the treatment of dissolved selenium in either neutral runoff from agricultural lands or neutral to acidic MIW from both coal and metal mining. Both active and passive treatment technologies are considered. Selenium bioreactor design principles are similar to those developed for sulfate reducing bioreactors commonly used for the treatment of high concentration metal-laden acid drainages.

**Additional Keywords:** Sulfate reduction; passive treatment; agricultural runoff

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## Introduction

The Kesterson National Wildlife Refuge in California was the focus of attention over 25 years ago when excessive selenium concentrations in the surface water were found to be the primary cause of mortality in fish and birds living there (National Research Council, 1989). The issue associated with mining is not new either. Selenium uptake in plants grown on reclaimed mine lands was reported at the 1988 American Society of Mining and Reclamation (ASMR), US Bureau of Mines and Office of Surface Mining Joint Conference on Mine Drainage held in Pittsburgh. Not many years later, a full session was dedicated to selenium at the 1995 ASMR Conference in Gillette, WY. A scan of combined ASMR, International Conference on Acid Rock Drainage (ICARD) and the West Virginia Surface Mine Drainage Task Force proceedings since 1980 yielded over 50 papers that addressed selenium issues.

The uses and natural occurrences of selenium are varied; they include:

- Paint, pigments, dye formulating
- Electronics
- Glass manufacturing
- Insecticide production
- Pulp and paper
- Ash piles, FGD blowdown
- Coal/oil combustion
- Agricultural water
- Petroleum processing
- Mining operations

## What’s the Problem?

The toxicity and health impacts of selenium in the natural and human environment is well established, although specific levels considered to represent a threat to human health or the environment are not held in universal agreement. Its bioavailability and toxicity is a function of oxidation state. With respect to human health, it is ironic that selenium is an essential trace nutrient found in health food stores, but that it also has a regulatory limit in drinking water.. The range of toxicity and nutritional deficiency is narrow, however. Unfortunately, selenium is bioaccumulative; its concentration may increase in organisms at successively higher levels in the food chain. The main issues associated with this enigmatic element include:

- It’s an aquatic life hazard
    - 1983 – Kesterson National Wildlife Refuge - California
    - Birth defects/death of birds, small animals, fish
- It’s a regulated drinking water contaminant
- Selenium cycle is not well understood

    - Uncertainty on bioavailability
    - Even if bio-available – what is a toxic threshold concentration?
- Often times low-concentration/high-volume flows make treatment expensive

The primary goal of any treatment process is to meet applicable standards. This is a controversial and complex topic and beyond the scope of this paper. The diversity in numeric standards is reflected in the list below.

- Freshwater aquatic life - 5 µg/L
- Primary Drinking Water Standard (DWS) Maximum Contaminant Limit (MCL) - 50 µg/L
- U.S. Fish and Wildlife Service has recommended 2 µg/L to protect fish, waterfowl and endangered aquatic species

Suffice it to say that any treatment process that reduces selenium loading to achieve or approach site-specific standards, whether concentrations are measured in the water itself or in fish tissue, is worthy of consideration. A secondary goal is to achieve dissolved selenium reductions as inexpensively as possible.

## Basic Selenium Geochemistry

The basic geochemistry of selenium needs to be briefly addressed before treatment alternatives can be considered. In contrast to sulfur, which is found in the same group in the periodic table, selenium has four primary oxidation states, including elemental selenium:

- elemental selenium ($Se_0$)
- -2 selenide ($Se^{-2}$), analogous to sulfide ($S^{-2}$)
- +4 selenite ($HSeO_3^-$ and $SeO_3^-$)
- +6 selenate ($SeO_4^{-2}$) analogous to sulfate ($SO_4^{-2}$)

The focus of this paper is mitigating mining impacts, so it is appropriate to focus on how selenium occurs in mining/geological environments. Ryser, et al. (2005) investigated the selenium-bearing minerals in the Western US Phosphate Resource Area in southeast Idaho and adjacent states (Utah and Wyoming) using micro-spectroscopic techniques. The researchers took great pains in statistically analyzing samples of a selenium-bearing shale interburden sandwiched between two major phosphate ore zones in the Permian-age Phosphoria Formation. The ore itself is virtually selenium-free. The research included test calibrations of various known selenium-bearing material standards, including two forms of elemental selenium (monoclinic and orthorhombic crystal forms). In their background research, Ryser et al. found that the most commonly reported forms of selenium in shale are selenium-substituted pyrite or precipitated metal selenides. From the testing on the Phosphoria Formation shale interburden, they concluded that there were three distinct mineral phases that contain selenium:

- Dzharkenite ($FeSe_2$) - an iron selenide analogous to pyrite
- An organic diselenide,
- Selenium-substituted pyrite.

Ryser et al. further observed that:

> *In all of these phases Se existed in the negative oxidation states, indicating that the oxidized Se present in weathered shales is a result of oxidative weathering of the reduced mineral Se to elemental Se, selenite, or selenate.*

In other words, the iron selenides in the overburden are analogous to pyrite in generating selenium-MIW. The noted absence of elemental selenium in the un-mined/un-weathered shale has implications regarding passive treatment in biochemical reactors which is subsequently discussed.

## Past and Present Selenium Treatment – USEPA BDATs

The precipitation of selenium from the dissolved state in MIW does not appear to be driven by equilibrium principles; rather, processes are governed by oxidation-reduction (Redox) conditions, biological activity, and sorption (MSE, 2001). The following processes with their associated MIW constraining conditions have been embraced by the USEPA as Best Demonstrated Available Technologies (BDAT) for selenium removal (MSE, 2001):

- Ferric coagulation/filtration
    - Typically for pH <7
    - Relies on a co-precipitation effect
    - Effective removal requires reduction of selenate to selenite
    - Performance can be compromised if arsenic is present
- Lime softening

- Reverse osmosis
    - Non-preferential process – all dissolved species present are removed
    - Pretreatment due to other typical mine water issues (i.e. scaling, fouling) may be required
- Electrodialysis
- Alumina
    - Selenite is adsorbed at pH range of 3 – 8
    - Silica can interfere at pH >4
    - Selenate adsorption is poor
- Ion exchange
    - Oxidized divalent selenate is needed
    - Competing ion effects can hinder effectiveness
    - Some specialty resins were tested
- Ferrihydrite precipitation with concurrent adsorption of selenium on the ferrihydrite surface
    - For adsorption – ferric ion ($Fe^{+3}$) needs to be present
    - Most effective removal occurs at pH 4-6
    - Somewhat effective up to pH 8
    - Phosphate, silicate, arsenic, carbonate can interfere

From 1999 to 2001, the USEPA and the US Department of Energy (DOE) co-sponsored research conducted by the Mine Waste Technology Program (MSE, 2001). This effort focused on demonstrating several selenium treatment/removal methods. Three technologies were tested in the field:

- Ferrihydrite Adsorption (baseline)
- Catalyzed Cementation
- Biological Reduction

A fourth technology was tested at a bench scale, Enzymatic Reduction.

The program objective was to practically treat to a selenium concentration of less than 50 µg/L (the Primary Drinking Water Standard). The research was conducted on MIW from the Kennecott

Utah Copper Corporation's Garfield Wetlands-Kessler Springs site, which had the following typical chemistry:

- <50 to >10,000 µg/L Se (up to 10 mg/L)
- 95%+ selenate
- TDS 1,000 - 5,000 mg/L

The following results were reported.

***Ferrihydrite*** - Ferrihydrite did not work on a consistent basis despite using various iron types, concentrations, and Fe:Se ratios. The process could achieve the program objective but at prohibitive reagent consumption and with questions on the TCLP stability of the resulting sludge.

***Catalyzed cementation*** – This was a process originally developed for arsenic, selenium, thallium removal; it removes metals by cementation on the surface of iron particles. It was believed to work on both selenite and selenate. Using proprietary catalysts, bench test work yielded favorable results but it did not work on a consistent basis during field trials.

***Biological Reduction (BSeR™)*** – This process used anaerobic solids bed reactors in which selenium was believed to be reduced to elemental selenium by biofilms and proprietary microorganisms; molasses was used as carbon source. The process consistently met the program objective; over 70% of samples had concentrations less than the detection limit (2 µg/L). The comparative economics (Table 1) of the three processes suggested that biological methods hold great promise.

Table 1 – Economic Comparison of Selenium Removal Technologies (from MSE, 2001)

| | BDAT | Cementation | BSeR™ |
|---|---|---|---|
| Capital | $1.0M | $1.1M | $0.6M |
| O&M | $2.1M | $1.2M | $0.14M |
| NPV | $17M | $9.5M | $1.1M |
| $/cubic meter | $3.67 | $2.16 | $0.35 |
| $/1,000 gal | $13.90 | $8.17 | $1.32 |
| $/kg selenium | $1,836 | $1,079 | $174 |

Based on 300 gpm plant, 2 mg/L selenium influent
2001 dollars; depreciation, leases, salvage and taxes were not considered

For comparison, the 2005 price quoted for elemental selenium metal was about $110/kg.

## Other Processes – Nanofiltration

Nanofiltration technology was evaluated by the US Geological Survey in 1996 for selenium removal from agricultural drainage. The test work showed that the technology achieved better selenate removal than selenite, which was not surprising since the process was designed for divalent rather than monovalent ions. Regardless, the system yielded 95+ % removal at selenium concentrations of less than 1,000 µg/L; membrane scaling would be an issue in high sulfate MIW sources.

## Summary Problems with Past & Present Processes

Collectively, the active treatment processes considered to date are typically non-selective for selenium; their implementation generates large amounts of secondary waste using a diverse array of reagents. While the processes are typically appropriate for the bulk removal of selenium in the presence of other metals, they have difficultly consistently producing clean effluents with less than 10 µg/L selenium. Of the processes considered, the biological processes appeared to provide the greatest promise in achieving effluent goals at a reasonable cost. A focused discussion of biological processes follows.

## Biological Reduction - General

The first ASMR paper addressing biological treatment for selenium was presented in 1994 (Wildeman, et al.). Selenium was listed as one of the constituents of concern in a neutral MIW tailings underdrain solution and an acidic MIW from a waste rock dump at a gold mine in Nevada. Earlier work by Gerhart and Oswald (1990) utilized “microalgae-bacterial” treatment of agricultural water in California.

A system in the Panoche Drainage District in the San Joaquin Valley is indicative of the potential success of biological systems (Fisher, 2004). Water chemistry in this locale ranges from 74 to 1,400 µg/L due to selenium rich soils; selenate is the primary species present. Numerous bioremediation studies in this area examined Algal-Bacterial Selenium Removal (ASBR) and Anoxic ponds which reportedly reduced selenate to selenite to elemental selenium and settled the resulting suspended precipitates. These systems typically exhibited removal efficiencies of about 80%.

Additional California-based research (Fisher, 2004) evaluated the behavior of constructed wetlands in which nine plant species were tested. The results showed 63% to 71% removal efficiencies for water containing about 20 µg/L selenium. The effluent contained from 3 to 6 µg/L Se. While the technology worked, the hydraulic retention times (HRTs) were on the order of several days.

Thus, the passive treatment concept for selenium is not new. The process is attractive because microbes degrade/transform the contaminant for a number of reasons, including:

- Selenium is used as a bacterial energy source
- Bacterial selenium removal is a detoxification mechanism
- Selenium behaves or resembles another ion (sulfate)
- Combinations of the above

Anaerobic biochemical reactors (BCR's) are especially attractive because of their ability to produce elemental selenium which is biologically unavailable at typical removal efficiencies of greater than 90%. There is some concern that nitrate and sulfate presence may interfere in these types of systems due to bacterial competition. It is interesting that while selenium speciation in bioreactors suggests the formation of elemental selenium, natural rock formations (at least in the context of phosphate deposits) do not appear to exhibit this species naturally. The implications of this observation are uncertain; perhaps the thermodynamics or kinetics of selenium sequestration in natural anaerobic environments favors organic complexation and co-precipitation with ubiquitous iron over the elemental form. This may be a subject suitable for future research.

This pioneering work resulted in more-selective selenium microbes being isolated; this was accompanied by advances in fixed film/biofilm media and a better understanding of operating conditions. The net result was that HRT's have been reduced from days to hours for active systems. Coincident advances were also made in passive technology.

The ABMet® Process is offered by the GE Water and Process Technologies group as a variant on the BSeR™ Process evaluated by MSE for the EPA/DOE. It has been used on several flue gas desulfurization projects at commercial scales with selenium levels ranging from 3,000 to 5,000 µg/L in the presence of up to 20,000 mg/L chloride. Removal efficiencies on the order of 98% to

99% removal are projected coincident with effluent concentrations as low as 10 µg/L (Sonstegard 2007).

## Historical Experience in Selenium Passive Treatment

### Definition of Passive Treatment

As discussed above, there appear to be several promising technologies for biologically treating selenium-bearing MIW. To properly focus the discussion, the following definition of passive treatment was proposed by Gusek (2002):

> ***Passive treatment*** is a process of sequentially removing metals and/or acidity in a natural-looking, man-made bio-system that capitalizes on ecological and geochemical reactions. The process requires no power and no chemicals after construction and lasts for decades with minimal human help.

It is a ***sequential*** process because no single treatment cell type works in every situation or with every MIW geochemistry. It is an ***ecological/geochemical*** process because most of the reactions (with the exception of limestone dissolution) that occur in passive treatment systems are biologically assisted. Lastly, it is a ***removal*** process because the system must involve the filtration or immobilization of the metal precipitates that are formed. Otherwise, they would be flushed out of the system, and the degree of water quality improvement would be compromised.

A truly passive system should also function for many years, without a major retrofit to replenish construction materials, and be able to function without using electrical power. Benning and Ott (1997) described a volunteer passive system outside of an abandoned lead-zinc mine in Ireland that has apparently been functioning unattended for over 120 years. Ideally, a passive treatment system should be designed to last for at least several decades.

### Early Passive Treatment Studies of Selenium-Bearing Mining Influenced Water

Wildeman, et al. (1994)

Wildeman, Filipek, and Gusek presented the results of "proof-of-principle" studies of passive treatment of neutral/alkaline cyanide mill tailing and acidic MIW sources at an undisclosed gold mine in Nevada at the 1994 ICARD Conference in Pittsburgh. This study was primarily focused on the selection of the preferred removal methods (aerobic or anaerobic/BCR) for addressing the parameters of concern for each MIW source; both sources contained selenium. The tailings derived MIW and the acidic MIW from a waste rock dump contained about 200 µg/L and 1,000 µg/L total (unfiltered) selenium, respectively. The results of the test demonstrated in principle that selenium concentrations could be decreased to less than the 10µg/L detection limit with both aerobic and anaerobic geochemical environments.

The proof-of-principle laboratory scale tests were the basis of building two pilot-scale passive treatment units:

- An algae-filled, multi-celled free water surface pond system with cascades for the tailing MIW, and
- A down-flow biochemical reactor and aerobic polishing wetland (in series) which received the acidic MIW from a pre-existing intermediate holding pond.

Performance data from these two systems has not been previously published. Table 2 below summarizes their respective performance with respect to selenium removal.

Table 2 – Passive Treatment Results – Nevada Gold Mine Tailing & Waste Rock MIW

| Pilot System | Flow Rate (gpm) | pH | Influent Se (µg/L) | Effluent Se (µg/L) | Percent Removal |
|---|---|---|---|---|---|
| Tailings MIW (Aerobic) | 10 | 7.5 | 40 | 16 | 60% |
| Waste Rock MIW (BCR) | 6 | 2.7 | 22 | <5 | ≥78% |

<u>Brewer Mine (1993-1995)</u>

The Brewer Mine is a closed gold mine in South Carolina with heap leach pad runoff and pit lake derived acidic MIW. The results of an 18-month study of a four-liter per minute capacity pilot scale biochemical reactor were described in Gusek, 2000. The primary parameters of concern were depressed pH, and elevated aluminum, iron, copper, and zinc. Several sampling events

Figure 1 – Brewer Pilot BCR

that included a longer list of analytes yielded data on of the pilot cell's performance with respect to selenium removal. Typical MIW selenium concentration were about 1,500 µg/L; BCR effluent selenium concentrations in the final sampling event were about 50 µg/L, a removal efficiency of about 96.7%. The influent sulfate concentration in this final sampling event was 5,200 mg/L; the field pH was 2.0.

**Recent Passive Treatment Studies**

Pahler, et al (2007)

Pahler, Walker, Rutkowski, and Gusek (2007) described a bench scale BCR study conducted at a gravel pit adjacent to the Colorado River in western Grand Junction, Colorado. The selenium concentrations in a mine pit dewatering trench ranged from 31 to 93 µg/L during the year prior to their study (Kerr, 2006); during the study, the influent concentration was about 20 µg/L, but it did spike to 70 µg/L during the five-month test. Background concentrations of sulfate ranged between 1,000 to 2,000 mg/L.

Four bench BCRs were constructed using 208-liter (55 gallon) polyethylene drums in 2006. Each was filled with varying amounts of homogenously-mixed sawdust, hay, wood chips, agricultural limestone, zero valent iron (ZVI) powder, and manure (Table 3). In the hope of inoculating the BCRs with bacteria acclimated to selenium, the manure was derived from cattle grazing in pasture areas known to have in selenium-rich soils. ZVI was included in varying amounts in order to determine if chemical reduction of selenium could enhance the expected biological reduction of selenium. Reactor 1, lacking ZVI, served as a baseline. Reactors 2, 3, and 4 had increasingly larger weight percentages of ZVI.

Table 3. BCR Media Compositions

| Component | Proportion of Each Component by Weight | | | |
|---|---|---|---|---|
| | Reactor 1 | Reactor 2 | Reactor 3 | Reactor 4 |
| Sawdust | 30% | 30% | 20% | 2.5% |
| Hay | 10% | 10% | 10% | 0% |
| Wood chips | 30% | 30% | 20% | 2.5% |
| Agricultural limestone | 20% | 5% | 5% | 5% |
| Zero valent iron | 0% | 15% | 35% | 85% |
| Cow manure | 10% | 10% | 10% | 5% |

Additional details on the conduct of the experiment are provided in the Pahler, et al. paper. The four reactors exhibited selenium removal efficiencies of up to 98%; effluent typically exhibited less than 2 µg/L selenium. In this situation, the ZVI incorporated cells offered no advantage over the baseline BCR which was ZVI-free.

The positive results of the bench scale BCR tests were the basis for designing a pilot scale BCR whose construction is planned for May, 2008. The pilot BCR will be sized to treat 4 L/min (one gpm). At full scale, the treatment cost associated with a BCR of similar design is estimated to be \$0.07 per $m^3$ of water treated (\$0.26 per 1,000 gallons) or \$115 per kilogram of selenium removed. At the 2005 selenium price quoted of \$115 per kg, and assuming a nominal cost for recovering a portion of the selenium resource, the treatment economics for this technology appear to be more favorable than active treatment

In the author's experience, the design of a selenium reducing BCR is similar to that required for a sulfate reducing BCR. Both are typically sized based on metal loading to a point as long as a minimum hydraulic detention time (HRT) is satisfied. The selenium BCR organic substrate may be enhanced with a sacrificial metal if the untreated MIW is lacking in metal (e.g., iron) concentration that would encourage the precipitation of iron selenide (see Ryser et al. 2005).

## Recent Experience in Active Selenium Treatment

A Western US metal mine waste rock leachate required treatment for reduction of TDS and selenium prior to discharge to surface water. The MIW at this site is comparatively dilute with respect to selenium; the influent concentration is typically about 30 µg/L while the effluent goal is 10 µg/L. The MIW has neutral pH with calcium, magnesium, and sulfate comprising the TDS with concentrations from approximately 5,000 mg/L to 8,000 mg/L. The other significant challenge of the project was the space constraints for the treatment system and the flow rate which ranged from 70 to 700 gpm with surge events in excess of 2,000 gpm. The treatment system, based on the results of seven months of pilot testing, includes a series of reverse osmosis (RO) units to provide initial volume reduction. The selenium reduction is achieved by biological treatment of the RO brine. Since the feed to the bio-treatment system is RO brine, the actual selenium concentration in the bioreactor feed is approximately 70 µg/L in a 16,000 mg/L TDS background. The bio-treated brine is then recombined with permeate from the RO to the TDS discharge limit and the combined stream is discharged to a nearby creek. Excess bio-treated brine is discharged to the sewer under a Pretreatment Discharge Permit. A process flow diagram of the system is provided in Figure 2.

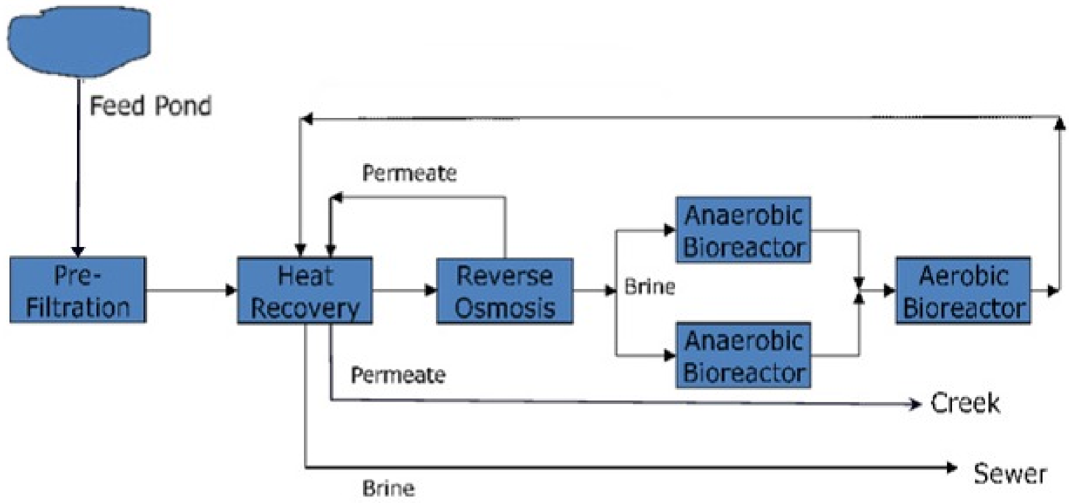

Figure 2 - Process Flow Diagram Active Selenium Bio-treatment at a Western US Metal Mine

The selenium bio-treatment process was developed on the bench-scale and then pilot scale including developing the culture which provided the selenium reduction in a high TDS environment. The culture was developed from onsite sediments and sediment from the Great Salt Lake. The bio-treatment system, fueled by molasses supported by phosphate and urea nutrient amendments, is currently operating on a feed with approximately 16,000 mg/L TDS. Sulfide generation has occasionally been an issue in operation.

Figure 3 Full scale biological treatment system

As of early 2008, the full scale system (Figure 3) has been operating for about two years and compliant water at less than 10 µg/L is being produced. The entire treatment facility covers about one acre.

The treatment cost is estimated to be about \$1.50 per $m^3$ (\$5.71 per thousand gallons) of MIW; this is slightly more than, but consistent with, the economics for the BSeR$^{TM}$ process cited in Table 1. However, because of the dilute MIW at this site (30 µg/L Se), the unit cost of selenium removal is estimated to be about \$50,000 per kilogram. About 12 kg of selenium is removed annually at this site.

## Conclusions

The treatment of selenium either actively or passively with removal efficiencies in the high ninety percent level has been demonstrated on a variety of MIW types, ranging from dilute, neutral pH to concentrated and acidic. The economics of passive treatment appear favorable if land area is available at a reasonable cost. Resource recovery may be able to offset some of the costs of treatment but only if the original MIW has a reasonably elevated selenium concentration; the "unit mass" cost of treating selenium-dilute MIW is not conducive to realizing resource recovery "credits".

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