



Treatment of coal mine leachate for neutralization and metal removal

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Abstract

Acid mine water poses a serious environmental risk due to its high toxicity and the potential for groundwater contamination. The study introduces an innovative process configuration and reactor design in order to minimize capital cost and gypsum scaling. A feasibility study on mine water from Khwezela Colliery in Mpumalanga province (South Africa) indicates that the designed process can remove H_3O^+ , Fe^{2+} , Fe^{3+} and Al^{3+} with CaCO_3 and Mn^{2+} and other metals with $\text{Ca}(\text{OH})_2$ at a cost of ZAR 6.89/m³. If only $\text{Ca}(\text{OH})_2$ is used, the cost is ZAR 16.25/m³.

Keywords: Acid mine water, metal removal, magnetite, calcium carbonate, reactor design

Introduction

In water strained South Africa (SA), mine water needs to be desalinated before it can be discharged into public streams (Mey & van Niekerk, 2009). Acidic mine drainage (AMD) resulting from mining activities is a major environmental concern as it requires neutralisation and desalination. Sludges generated during neutralisation are rich in metals such as Fe^{2+} , Fe^{3+} , Al^{3+} and Mn^{2+} and must be handled as toxic waste (Maree, et al., 2013).

The High-Density Sludge (HDS) process was developed by the Bethlehem Steel Corporation (USA) for the neutralization of acid mine water. In this process, a part of the settled sludge is mixed with the AMD, followed by the addition of lime. Due to the “seeding” effect, a denser sludge is produced (Kostenbader & Haines, 1970). The widely used HDS process was optimized by Osuchowski (Osuchowski, 1992). The HDS process consists of the following stages: (i) pH correction/sludge conditioning; (ii) Neutralization/aeration, and (iii) Solid/liquid separation. Addition of lime in the pH correction stage results in the final treated water having a pH of around 8, and aeration resulted in Fe^{2+} -oxidation. Due to the high

price of $\text{Ca}(\text{OH})_2$ (ZAR 3 950/t), CaCO_3 has been used for neutralization at Navigation instead of the HDS process (Günther, et al., 2003).

In Mpumalanga, three mine water desalination plants are in operation, namely Emalahleni (50 ML/d), Optimum (25 ML/d) and Witbank South (25 ML/d) (Mogashane, 2022). The plant at Emalahleni was designed to treat 50 000 m³/d of mine wastewater and was built at a cost of R900 million (R18 million/(1000 m³/d). Khwezela Colliery needs to treat 21 ML/d of acid mine water, called Pit water, in order to lower the water level to get access to the coal seam. This water must be neutralized and stabilised with respect to gypsum crystallization before it can be desalinated in the Emalahleni Water Reclamation Plant. The Pit water contains 612 mg/L iron, 5 270 mg/L sulfate, 3 643 mg/L acidity (as CaCO_3), and has a pH of 3.1.

A limestone neutralization plant with a capacity of 0.5 ML/d (20.8 m³/h) is available for treatment (Günther, et al., 2003; Maree & du Plessis, 1994; Maree, et al., 1999). The plant (two-stage completely-mixed reactor followed by a clarifier with sludge recirculation; 2 h residence time) use precipitated calcium carbonate for neutralization (ZAR 1 050/t).

It avoids iron(II)-oxidation. In the case of CaCO_3 , Iron(II)-oxidation needs to be carried out at pH 5, where the rate of iron(II)-oxidation is slow. This slow rate of oxidation was overcome by providing a high suspended solids concentration of 50 g/L through sludge recirculation. The following shortcomings were experienced with the current neutralization plant: (i) limited capacity of 0.5 ML/d while 21 ML/d needs to be treated; (ii) gypsum scaling caused pipe blockages and built-up of gypsum at the floor of the clarifier, resulting in regular breaking of the gearbox; (iii) high capital cost of ZAR 9 million for a plant with a capacity of 1 ML/d. The capacity of this plant needs to be increased to 21 ML/d within a short space of time and at lowest cost.

Maree (UNISA/ROC Water Technologies) designed a process configuration that will meet the following requirements: (i) low running cost by using CaCO_3 due to its low price compared to that of Ca(OH)_2 ; (ii) iron(II)-removal when CaCO_3 and Ca(OH)_2 is used for neutralization; (iii) gypsum crystallization to its saturation; (iv) minimize capital cost needed for solids separation (Maree, 2024). The purpose of this investigation was to evaluate the UNISA/ROC Water design and to optimize it.

Objectives

The following objectives were set for the project: (i) provide a process configuration for the treatment of 21 ML/d acid mine water (ii) predict the water quality of the treated water (iii) provide a process configuration and reactor design that will minimize running and construction cost and minimize maintenance due to gypsum scaling and (iv) determine the feasibility of the process.

Materials and Methods

Feedstock. Pit water from Khwezela Colliery, Mpumalanga, South Africa, was used for the design of a neutralization plant.

Procedure. OLI software simulations were used to predict the water quality when the pH was increased from 2.8 to 8.2 using (i) CaCO_3 , for raising the pH to 4.5, followed by (ii) Ca(OH)_2 , for raising the pH to 8.2. The simulation showed the compounds removed

due to metal precipitation and gypsum crystallization.

Experimental. The effect of the following parameters were investigated: (i) alkali selection (CaCO_3 , Ca(OH)_2) and (ii) process configuration (CaCO_3 combined with Ca(OH)_2 , and Ca(OH)_2 alone).

Analytical. Standard procedures were used to collect samples at various phases, filter them (Whatman No. 1), and measure their concentrations of Fe(II), Fe(III), pH, Ca, and alkalinity (APHA, 2012). Metals were analyzed using inductively coupled plasma-atomic emission spectroscopy (ICP-OES) (iCAP 7000 Series, ANATECH, South Africa). The pH/EC meters were calibrated before the start of each set of experiments and during the experiment using calibration buffers.

OLI software simulations. The OLI ESP software program was used to predict the behavior of metals dissolved in water during dosing of alkalis, like CaCO_3 and Ca(OH)_2 . The solubility of CaSO_4 and metal hydroxides were identified as a function of temperature and concentration.

Results and Discussion

Water quality

Iron is the main dissolved component of Pit water and is split between Fe^{2+} (212 mg/L) and Fe^{3+} (400 mg/L). The Fe^{2+} and Fe^{3+} fractions represent 612 mg/L of the total acidity of 3 643 mg/L. Dissolved Al^{3+} is the second largest contributor to the acidity, namely 260 mg/L (as Al) (or mg/L as CaCO_3).

Table 1 shows the behaviour of Pit water when treated with CaCO_3 , as predicted with OLI simulations. For Fe^{3+} to be in solution, the water needs to contain 282 mg/L acid (as H_3O^+). The following observations were made when the CaCO_3 dosage was increased stepwise from 0 to 4 521 mg/L: (i) Free acid (H_3O^+) (282.8 mg/L) and Fe^{3+} (399.9 mg/L) were removed gradually as the pH was increased from 2.0 to 3.9, at a dosage of 2 497 mg/L. The free acid was removed as CO_2 that appeared in gas and liquid phases. Fe^{3+} was removed as Fe(OH)_3 (Bernalite), (ii) Al^{3+} (259.4 mg/L) was removed as Al(OH)_3 (Gibbsite) in the pH 3.9 to 4 .8 at a CaCO_3 dosage of 525 mg/L, (iii) Fe^{2+} (212 mg/L) was

removed mainly as Fe_3O_4 (Magnetite) when the pH was raised to 5.3 by dosing a further 181 mg/L, and (iv) No Mn^{2+} and Mg^{2+} removal were obtained with CaCO_3 treatment.

Ca(OH)_2 can be used after CaCO_3 treatment for removal of Mn^{2+} and even Mg^{2+} . Only Ca(OH)_2 can be used for removal of all the metals, namely Fe^{2+} , Fe^{3+} , Al^{3+} , Mn^{2+} and Mg^{2+} . Table 2 shows the behaviour of Pit water when treated with Ca(OH)_2 , as predicted with OLI simulations. The following observation were made: (i) lime removed Fe^{3+} and Al^{3+} in the same way as in the case of CaCO_3 , first Fe^{3+} at pH below 4, and thereafter Al^{3+} ; (ii) Fe^{2+} , in the absence of O_2 , and in the presence of Fe^{3+} , was removed as Fe_3O_4 (Magnetite), at pH values 4.8 and higher, for both CaCO_3 and Ca(OH)_2 ; (iii) Mn^{2+} was removed as Mn(OH)_2 (Pyrochrochrite) at pH 9.8; (iv) Mg^{2+} was removed as Mg(OH)_2 (Brucite) at pH 10.2, and (v) SO_4^{2-} was removed through $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ (Gypsum) crystallization to the saturation level.

The above findings allowed us to minimize alkali cost (Table 3). By using CaCO_3 for removal of H_3O^+ , Fe^{2+} , Fe^{3+} and Al^{3+} and Ca(OH)_2 for removal of Mn^{2+} and other metals amounted to R6.89/ m^3 and by using Ca(OH)_2 for removal of H_3O^+ , Fe^{2+} , Fe^{3+} , Al^{3+} , Mn^{2+} and other metals amount to R16.25/ m^3 .

Table 3 Alkali cost for different treatment options

Parameter	Option	
	$\text{CaCO}_3/\text{Ca(OH)}_2$	Only Ca(OH)_2
CaCO_3 dosage (mg/L)	3 703.0	
Ca(OH)_2 dosage (mg/L)	4 98.0	3 496.0
CaCO_3 cost (R/ m^3)	4.57	
Ca(OH)_2 cost (R/ m^3)	2.31	16.25
Total cost (R/ m^3)	6.89	16.25
Cost ratio	42.4	100.0
Total cost (R/month)	4 397 637	10 371 522
Notes:		
CaCO_3 purity (%)	85.0	
Ca(OH)_2 purity (%)	85.0	
CaCO_3 price (ZAR/t)	1 050.0	
Ca(OH)_2 price (ZAR/t)	3 950.0	
Flow (ML/d)	21.0	

Process configuration

The process configuration made provision for neutralization with precipitated calcium carbonate and/or lime, followed by solids separation. For neutralization, an innovative reactor design is proposed that will meet the following requirements: (i) achieve iron(II)-removal while CaCO_3 is used as the alkali, (ii) minimize gypsum scaling by integration of the completely-mix reactor with the clarifier, and (iii) eliminate the need for a clarifier by passing the neutralized water on directly to the slimes dam, where solid waste is disposed. The following process configuration will meet the mentioned requirements: (i) CaCO_3 neutralization at pH 4.8 in a complete-mix reactor to achieve Fe(OH)_3 , Al(OH)_3 , Fe_3O_4 precipitation, $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ crystallization, and sludge separation in a clarifier (directly above the complete-mix reactor), (ii) Ca(OH)_2 treatment at pH to 9.2 to precipitate Mn(OH)_2 and other metals, together with aeration to allow for CO_2 stripping and oxidation of Mn(OH)_2 to MnO_2 . The rate of Fe_3O_4 formation needs to be confirmed in the laboratory. If the rate of Fe_3O_4 formation is slow, Fe^{2+} will be oxidised through aeration. The cost of aeration will be included in the feasibility section. The removal of Fe^{2+} through formation of magnetite (Fe_3O_4) is an

Table 4 Comparison between feasibility of conventional and proposed treatment

Parameter	Proposed	Conventional
Capital cost (R/(ML/d))	1 809 368	9 000 000
Capital cost (R)	37 996 725	797 931 216
Capital redemption cost	0.79	3.91
Chemical cost (R/ m^3)	6.89	16.25
Electricity (kW/(ML/d))	8.38	8.38
Electricity price (ZAR/kWh)	2.00	2.00
Electricity (kWh/ m^3)	0.60	1.00
Electricity (ZAR/ m^3)	1.20	2.00
Labour (20 labourers; R10 000/month)	0.31	0.31
Project management, Admin, Assurance (R100 000/m)	0.16	0.16
Total (R/month)	9.34	22.63
Ratio	41.30	100.00

innovative step that will be protected through registration of a provisional patent (Maree, 2024), and (iii) Solids removal and disposal in a slimes dam.

Reactor design

The capital cost for neutralization with a conventional design amount to R 9 million for a plant with a capacity of 1 ML/d. The conventional design makes provision for a completely mixed reactor for sludge conditioning with $\text{Ca}(\text{OH})_2$, completely mixed reactor for contacting feed water with conditioned sludge, clarifier, sludge recirculation and sludge disposal.

Figure 1 shows the design that will allow for reduced capital cost. The complete-mixed reactor and clarifier is combined into a Reactor-Clarifier. This design will reduce the area where gypsum scaling could occur, as sludge is moved from the clarifier zone on top of the complete-mixed zone through a single opening (no recycle pump and sludge pipelines), to the complete-mixed zone. The stirrer needed for mixing was also used for transferring sludge from the clarifier zone to the complete mixed zone, by adding a second small impeller to the shaft of the mixer in the bottom opening of the clarifier. No expensive concrete structure is needed for the clarifier, only partition plates. For a flow of 21 ML/d, at a reaction time of 2 h, 2 square units will be constructed, each unit will have a side length of 14.8 m and a height of 4 m. Each unit will be split into 4 sections, each section having its own mixer. The plates, that separate the settling zone from the complete-mixed zone, will have openings to allow for sludge recirculation. Settled sludge in the clarifier will be moved downwards through the opening due to the rotation of the small impeller. The same volume will be returned through the openings higher up in the separation plates. The large impeller at the bottom of the shaft will be used for mixing. The settling zone is equipped with openings at the top of the complete-mix zone to allow for air to escape.

Clarification of the treated water and sludge disposal is combined, by passing the treated water over a slimes dam where sludge is collected. A retention time of 24 h

is provided to allow sufficient time for sludge settling and compaction. The water height of 50 mm allows for good contact between the gypsum rich water and the settled gypsum on the floor, which will stimulate gypsum crystallization to its saturation level. For a flow rate of 21 ML/d, an area of 648×648 m will be needed. The slimes dam will be split into several compartments to allow for (i) filling (ii) reaction time and settling, and (iii) discharge of treated water.

Feasibility

Table 4 compares the total cost of the proposed process configuration (Option 1) with that of the conventional treatment (Option 2). The capital cost of the Reactor-Clarifier and Slimes dam is estimated at R 1.81 million for a plant with a capacity of 1 ML/d. The Capital Redemption cost was calculated for an interest of 10% per year and a payback period of 10 years (120 months). It shows the feasibility for removal of H_3O^+ , Fe^{2+} , Fe^{3+} , Al^{3+} Mn^{2+} and other metals. The feasibility was calculated from the Capital Redemption cost, chemical cost, electricity cost and labour. It was noted that the cost in the case of Option 1 amounted to ZAR 9.34/ m^3 compared to the cost of ZAR 22.63/ m^3 for Option 2.

Conclusions

The following conclusions were made: (i) CaCO_3 can be used for removal of Fe^{2+} , Fe^{3+} and Al^{3+} and lime for removal of Mn^{2+} and other metals at a cost of R6.89/ m^3 (3 000 mg/L CaCO_3 ; 300 mg/L $\text{Ca}(\text{OH})_2$, R 1 050/t CaCO_3 ; R 3 095/t; $\text{Ca}(\text{OH})_2$) (Option 1), (ii) as an alternative to Option 1, $\text{Ca}(\text{OH})_2$ alone can be used at a cost of R 16.25/ m^3 (3000 mg/L $\text{Ca}(\text{OH})_2$) (Option 2); (iii) the combined Reactor-Clarifier design, together with a slimes dam, can be used as an alternative to the conventional complete-mixed reactor/clarifier at reduced capital cost (R 1.91 million for a 1 ML/d plant versus R 9.0 million in the case of the conventional design); and (iv) the total cost of Option 1 (CaCO_3 and $\text{Ca}(\text{OH})_2$) and the new design will amount to R 9.34/ m^3 compared to R 22.63/ m^3 in the case of Option 2 ($\text{Ca}(\text{OH})_2$) alone and the conventional design.

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Table 1 Water quality of Pit water and after treatment with CaCO3 as predicted with OLI software

CaCO ₃ [mg/L]	0	1400	2.0	282.8	399.9	212.0	0.0	0.0	0.0	0.0	260.0	0.0	72.8	0.0	0.0	0.0	0.0	0.0	250.0	200.0	0	0.0	0.0	150.0	5937.0	100.0	0.0	0.0	0.0	995 417	
H ₂ SO ₄ [mg/L] (Y2)	1400	1400	2.0	249.1	283.2	211.9	223.3	0.0	0.0	0.0	260.0	0.0	72.8	0.0	0.0	0.0	0.0	0.0	0.0	249.9	400.1	0	0.0	0.0	150.0	5936.4	100.0	0.0	202.2	60.0	995 260
pH	1400	1400	2.1	212.5	171.8	211.9	436.4	0.0	0.0	0.0	260.0	0.0	72.8	0.0	0.0	0.0	0.0	0.0	0.0	249.9	539.3	262	0.0	0.0	150.0	5789.7	100.0	0.0	408.0	120.0	995 062
H ₃ O ⁺ 1 [mg/L] (Y2)	1400	1399	2.2	166.5	77.9	212.0	616.2	0.0	0.0	0.0	260.0	0.0	72.8	0.0	0.0	0.0	0.0	0.0	0.0	249.9	534.0	1145	0.0	0.0	150.0	5296.9	100.0	0.0	616.7	180.0	994 770
H ₂ SO ₄ [mg/L] (Y2)	1399	1399	2.4	98.3	15.9	212.0	734.9	0.0	0.0	0.0	260.0	0.0	72.8	0.0	0.0	0.0	0.0	0.0	0.0	250.0	528.3	2030	0.0	0.0	150.0	4803.2	100.0	0.0	827.5	240.0	994 494
	1398	1398	3.9	2.9	0.0	212.0	765.4	0.0	0.0	0.0	259.4	1.9	72.8	0.0	0.0	0.0	0.0	0.0	0.0	250.0	522.8	2915	0.0	0.0	150.0	4309.5	100.0	0.0	1035.0	300.1	994 247
	1398	1398	4.0	2.5	0.0	212.1	765.8	0.0	0.0	0.0	170.4	259.7	72.8	0.0	0.0	0.0	0.0	0.0	0.0	250.1	519.7	3791	0.0	0.0	150.1	3822.9	100.1	0.0	1247.1	360.3	994 314
	1357	1357	4.1	2.0	0.0	212.2	766.2	0.0	0.0	0.0	81.6	516.8	72.9	0.0	0.0	0.0	0.0	0.0	0.0	250.3	517.0	4668	0.0	0.0	150.2	3336.4	100.1	98.7	1406.7	405.9	964 829
	1233	1233	4.8	0.4	0.0	212.4	766.7	0.0	0.0	0.8	750.8	72.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	250.4	518.6	5527	0.0	0.0	150.3	2860.2	100.2	98.7	1406.9	416.4	876 567
	1152	1152	5.3	0.1	0.0	22.5	40.1	787.1	0.0	0.0	753.0	72.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	250.4	548.0	6261	0.0	0.0	150.2	2449.3	100.2	98.7	1413.9	438.4	819 079
	1266	1266	5.9	0.0	0.0	5.5	0.0	830.6	13.5	0.0	753.1	72.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	250.4	617.4	6825	0.0	0.0	150.3	2134.5	100.2	98.7	1413.6	549.6	900 000

Table 2 Water quality of Pit water and after treatment with Ca(OH)₂ as predicted with OLI software

Ca(OH) ₂ [mg/L]	H ₂ SO ₄ [mg/L] (Y2)	pH	H ₃ O ⁺ 1 [mg/L] (Y2)	Fe ⁽⁺³⁾ Liq1 [mg/L] (Y2)	Fe ⁽⁺²⁾ Liq1 [mg/L] (Y2)	Fe(OH) ₃ (Bernalite) - Sol [mg/L] (Y2)	Fe ₃ O ₄ (Magnetite) - Sol [mg/L] (Y2)	FeCO ₃ (Siderite) - Sol [mg/L] (Y2)	Al ⁽⁺³⁾ Liq1 [mg/L] (Y2)	Al(OH) ₃ (Gibbsite) - Sol [mg/L] (Y2)	Mn ⁽⁺²⁾ Liq1 [mg/L] (Y2)	Mn(OH) ₂ (Pyrochroite) - Sol [mg/L] (Y2)	Mg ⁽⁺²⁾ Liq1 [mg/L] (Y2)	Mg(OH) ₂ (Bricolite) - Sol [mg/L] (Y2)	Ca ⁽⁺²⁾ Liq1 [mg/L] (Y2)	CaSO ₄ ·2H ₂ O (Gypsum) [mg/L] (Y2)	CaCO ₃ (Calcite) - Sol [mg/L] (Y2)	Ca ₆ Al ₂ (SO ₄) ₃ (OH) ₁₂ ·26H ₂ O (Ettringite) - Sol [mg/L] (Y2)	Na ⁽⁺¹⁾ Liq1 [mg/L] (Y2)	SO ₄ ⁽⁻²⁾ Liq1 [mg/L] (Y2)	Cl ⁽⁻¹⁾ Liq1 [mg/L] (Y2)	H ₂ O ⁽⁻²⁾ [mg/L] (Y2)
0	1400	2.0	283	400	212	0	0	0	260	0	73	0	250	0	200	0	0	0	150	5937	100	995417
500	1400	2.0	237	242	212	302	0	0	260	0	73	0	250	0	470	0	0	0	150	5937	100	995289
1000	1399	2.1	183	103	212	568	0	0	260	0	73	0	250	0	538	874	0	0	150	5449	100	995027
1499	1399	2.4	94	14	212	739	0	0	260	0	73	0	250	0	531	2065	0	0	150	4785	100	994739
1998	1399	4.0	3	0	212	766	0	0	222	110	73	0	250	0	525	3255	0	0	150	4122	100	994621
2498	1399	4.1	2	0	212	766	0	0	101	460	73	0	250	0	521	4439	0	0	150	3466	100	994802
2998	1399	4.8	0	0	153	539	247	0	1	752	73	0	250	0	521	5608	0	0	150	2816	100	994858
3496	1399	9.8	0	0	0	0	847	0	0	753	4	111	186	155	545	6699	0	0	150	2223	100	994394
3994	1398	10.2	0	0	0	0	848	0	0	752	1	117	24	543	587	7655	0	0	150	1673	100	994021
4493	1398	11.4	0	0	0	0	848	0	5	516	0	118	0	601	634	7151	0	1790	150	1547	100	993877
4991	1398	11.4	0	0	0	0	849	0	5	164	0	118	0	602	634	5992	0	4625	151	1547	100	993715

Table 3 Alkali cost for different treatment options

Parameter	Option	
	CaCO ₃ / Ca(OH) ₂	Only Ca(OH) ₂
CaCO ₃ dosage (mg/L)	3 703.0	
Ca(OH) ₂ dosage (mg/L)	4 98.0	3 496.0
CaCO ₃ cost (R/m ³)	4.57	
Ca(OH) ₂ cost (R/m ³)	2.31	16.25
Total cost (R/m ³)	6.89	16.25
Cost ratio	42.4	100.0
Total cost (R/month)	4 397 637	10 371 522
Notes:		
CaCO ₃ purity (%)	85.0	
Ca(OH) ₂ purity (%)	85.0	
CaCO ₃ price (ZAR/t)	1 050.0	
Ca(OH) ₂ price (ZAR/t)	3 950.0	
Flow (ML/d)	21.0	

Table 4 Comparison between feasibility of conventional and proposed treatment

Parameter	Proposed	Conventional
Capital cost (R/(ML/d))	1 809 368	9 000 000
Capital cost (R)	37 996 725	797 931 216
Capital redemption cost	0.79	3.91
Chemical cost (R/m ³)	6.89	16.25
Electricity (kW/(ML/d))	8.38	8.38
Electricity price (ZAR/kWh)	2.00	2.00
Electricity (kWh/m ³)	0.60	1.00
Electricity (ZAR/m ³)	1.20	2.00
Labour (20 labourers; R10 000/month)	0.31	0.31
Project management, Admin, Assurance (R100 000/m)	0.16	0.16
Total (R/month)	9.34	22.63
Ratio	41.30	100.00

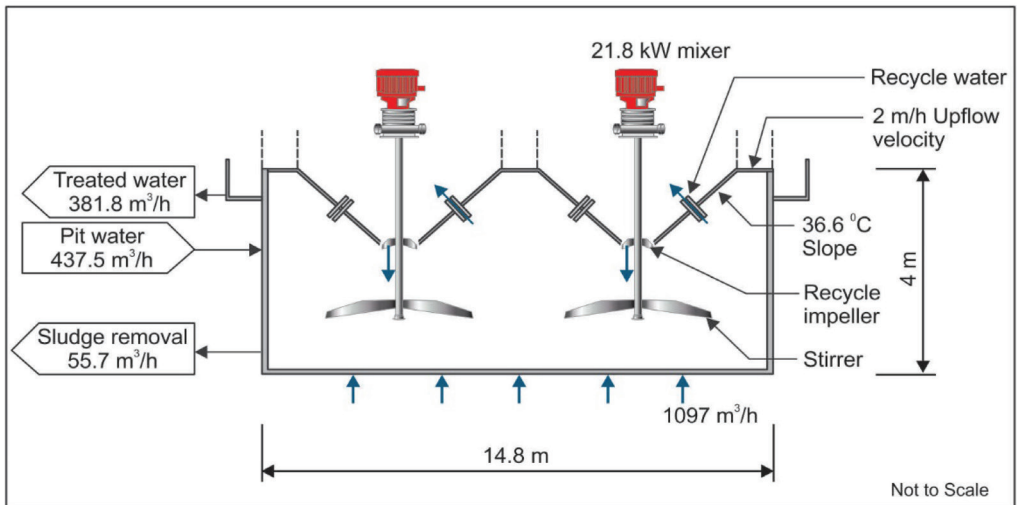


Figure 1 Reactor-Clarifier proposed for treatment