# Study and analysis of a full-scale laboratory successive alkalinity producing system for iron removal during the treatment of acid mine drainage

The Successive Alkalinity Producing System (SAPS) is widely accepted for the treatment of acid mine drainage (AMD). The effectiveness of SAPS depends upon many parameters such as hydraulic retention time (HRT), influent quality, characteristics of organic substrate and their design aspects etc. In this laboratory SAPS column study, four synthetic AMD solutions were treated for 1 day, 2 days, 4 days, 7 days and 10 days HRTs to determine the performances of SAPS using cow compost as organic substrate, saw dust and lime stone in parallel in identical conditions. In this research, studies were carried out to find the effect of hydraulic retention time on iron removal and alkalinity generation by laboratory SAPS. The current results reveal that 100% iron a removal has been found. Higher rate of alkalinity generation was observed in initial reaction period; then it got slower down after passage of time. The encouraging results obtained on performance of laboratory scale set up of AMD is useful for large scale implementation in mines for treating AMD influent mine water and for design of various component of SAPS for its optimum performance.

**Keyword:** Acid mine drainage; passive alkalinity generation; successive alkalinity producing system; HRT, ORP

#### 1. Introduction

The detrimental impact of mining activity on environment has been realised and considerable research is still going on (Patel and Patel, 2016). The mining of pyritic minerals produces sulphuric acid and metal ions (Skousen et al., 2017). The acid mine drainage (AMD) is produced by oxidation of sulphide-pyrite, marcasite and pyrrhotite (Equeenuddin et al., 2010). The AMD may have low pH, high electrical conductivity and high concentration of Fe, Al and Mn (Akcil et al., 2006). Following chemical reaction takes place during AMD formation:

$$\text{FeS}_2 + 3.5\text{O}_2 + \text{H}_2\text{O} \rightarrow \text{Fe}^{2+} + 2\text{SO}_4^{-2-} + 2\text{H}^+ \qquad \dots 1$$

$$Fe^{2+} + 0.25O_2 + H^+ \rightarrow Fe^{3+} + 0.5H_2O$$
 ... 2

$$\text{Fe}^{3+} + 3\text{H}_2\text{O} \rightarrow \text{Fe}(\text{OH})_3 + 3\text{H}^+ \qquad \dots 3$$

$$\text{FeS}_2 + 14\text{Fe}^{3+} + 8\text{H}_2\text{O} \rightarrow 15\text{Fe}^{2+} + 2\text{SO}_4^{-2-} + 16\text{H}^+ \quad \dots 4$$

AMD has very detrimental effect on both human as well as aquatic life and material. As a result proper treatment of AMD is essential before discharging into rivers (Neculita et al., 2008). AMD can be treated by two ways, one by active treatment and second by passive treatment process. The alkaline chemicals such as limestone and NaOH are used whereas in passive treatment system AMD is treated with the help of natural process. Passive treatment process is less costly and eco-friendly. Successive alkalinity producing system (SAPS) is one of the passive treatment systems which utilises the principle of anoxic limestone drain and anaerobic wetland system both. In SAPS AMD flows vertically through organic substrate layer than lime stone layer at bottom (Keplar et al., 1994). SAPS system can treat rich iron and aluminium containing AMD (Jage et al., 2000). The perforated pipe is provided at the bottom of SAPS due to which vertical flow is achieved (Ordonaz et al., 2012). The SAPS has been found successful in the treatment of AMD in South Korea (Bhattacharya et al., 2008). In this study four synthetic AMD has been evaluated for iron, aluminium and manganese removal along with alkalinity generation in similar condition.

#### 2. Methods and materials

#### 2.1 EXPERIMENTAL SET UP OF SAPS

Each SAPS unit consists of 3 components, first the AMD container placed at elevated position, second the SAPS cell in which the various materials such as limestone saw dust and cow compost were filled up as shown in Fig.1. Four PVC container of 80 liter capacity were chosen for making SAPS cells. At the bottom of the cell, a 2.5 cm thick gravel bed was filled up. At this portion, a perforated pipe was fitted for the

Messrs. M. D. Patel, R. K. Jade, and P. K. Dewangan, Associate Professor, Department of Mining Engineering, National Institute of Technology, Raipur 492010, India



Fig.1 Laboratory arrangement for SAPS column study (Patel et al., 2019)

purpose of flushing. The other end of bottom pipe was connected to oxidation cell. Above the layer of gravel, a limestone (>90% CaCO<sub>3</sub>) layer of 15 cm thickness was laid. A layer of 5 cm thick made up of saw dust and 22 cm thick cow compost layer were made above the limestone layer. 2.5 cm thick gravel was packed at the top and finally AMD water was allowed to fill up to 15 cm height above the top gravel pack. The estimation of quantity of limestone and organic substrate were done on the basis of recommendations of design guidelines by Cristopher Raymond Jage3. On this basis, the estimated quantity of limestone was approximately 27 kg and volumes of organic substrate were estimated at 29 liters approximately

A total of 4 synthetic AMD solutions were prepared with various compositions. All the experiments were performed in identical conditions with the temperature ranges from 21°C to 35°C in parallel four similar SAPS set up simultaneously in the same time period. The cow compost is taken from agricultural farm which is having sufficient population of bacteria. The cow compost in all the SAPS unit was kept same. A polyethylene net of 16 mesh was placed between different layers of materials to avoid the mixing and for maintaining the easily flow condition inside the SAPS cell and third unit is oxidation cell in which we allow the oxic conditions for water coming from the SAPS cell, were metal gets precipitated and pH increases. In each SAPS system, the oxidation cell of 50 liters volume was attached with 3 cm thick layer of limestone at the bottom. SAPS column experiments were conducted with four different synthetic AMD with the composition as shown in Table 1, considering major component of coal mine AMD as iron, aluminum and manganese, which are predominantly

found. The SAPS filled with cow compost, saw dust, limestone and normal water, then SAPS cells were left for 15 days as acclimation period for sufficient growth of bacteria. High concentration of hydrogen sulfide is produced after 15 days which is indicated by strong smell of rotten egg, confirming the large number of sulfate reducing bacteria (SRB) grown up in the SAPS cell. Growth of SRB is an essential condition for proper functioning of SAPS cell. After 15 days acclimation period. All the four synthetic AMD solutions were allowed to pour in the SAPS cell with control flow rate having different HRTs.

In this experiment four identical SAPS units having similar composition of limestone and organic substrate were operated simultaneously in each round (24 days), hence 4 synthetic

TABLE 1: COM	APOSITION OF AMD	USED IN THE	EXPERIMENTS
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	AMD A <sub>2</sub>	AMD B <sub>2</sub>	AMD C <sub>2</sub>	AMD D <sub>2</sub>
pН	4.30	3.60	2.72	2.50
ORP (mV)	105.2	107.30	108.40	106.30
DO (mg/L)	6.58	5.65	6.42	5.65
Total Fe (mg/L)	95.60	121.50	172.30	194.20
$Fe^{2+}$ (mg/L)	86.00	90.90	91.10	88.90
$Fe^{3+}$ (mg/L)	9.60	30.60	81.20	105.30
(Fe <sup>3+</sup> /Fe <sup>2+</sup> ) ratio	0.111	0.330	0.890	1.184
Al (mg/L)	40.00	40.00	40.00	40.00
Mn (mg/L)	15.00	15.00	15.00	15.00
Ca (mg/L)	125.00	125.00	125.00	125.00
Mg (mg/L)	100.00	100.00	100.00	100.00
SO <sub>4</sub> - (mg/L)	1044.00	1051.00	1052.00	1058.00
TDS (mg/L)	1321.20	1346.70	1443.80	1446.60

AMD solutions of different composition were treated. Synthetic AMD were prepared just before the commencement of experiment round. A blank column run for synthetic AMD was also conducted for 24 days in which no significant changes were noticed in characteristics of synthetic AMD. In every round, cow compost having the similar range of composition was loaded for the study. Five hydraulic retention time (HRTs) of 1d, 2d, 4d, 7d and 10d were proposed. Flow rate were maintained with the help of intravenous infusion set (I-V set).

#### 3. Results and discussion

In this phase four synthetic AMD of following composition were processed in SAPS cell

In the experiments, all the four SAPS cells (SAPS A, SAPS B, SAPS C and SAPS D) were prepared as described in previous chapter in similar and identical manners. After acclimation period the AMD A<sub>2</sub>, AMD B<sub>2</sub>, AMD C<sub>2</sub> and AMD D<sub>2</sub> were allowed to flow in SAPS A, SAPS B, SAPS C and SAPS D respectively as presented in Table 1. Then all the samples were collected at port P<sub>1</sub>, port P<sub>2</sub>, and port P<sub>3</sub> for different HRT's of 1d, 2d, 4d, 7d and 10 d respectively.

#### 3.1 SAPS PERFORMANCE WITH AMD A<sub>2</sub>

## 3.1.1 Oxidation reduction potential (ORP) and dissolved oxygen (DO) status in SAPS cell A

The ORP in SAPS cell A for AMD  $A_2$  showed downward trend at port  $P_3$  from 105.20 mV to - 212.30, -214.50, -311.20, -322.30 and -382.70 mV for 1d, 2d, 4d, 7d and 10 d HRTs respectively, which indicated that a strong reducing environment was prevailing inside the SAPS cell A. The negative ORP inside the SAPS is an essential condition for efficient operation of SAPS system as presented in Fig.2. The DO level in SAPS cell for AMD  $A_2$  was dropped from 6.58 to zero up to port  $P_2$  for all HRTs, which was an encouraging indicator of attainment of anaerobic conditions inside the SAPS cell. Similarly, DO levels were also reported as zero at port  $P_3$  for all the HRTs as given in Fig.3. Therefore DO and ORP changed status is favourable for anaerobic reducing environment inside all the four SAPS cells. The total dissolved solids (T.D.S.) and sulfate were also decreasing



Fig.2 ORP variation in SAPS with AMD A2



during SAPS process which confirmed the metal removals and alkalinity generation in SAPS cell.

#### 3.1.2 Iron Removal Behavior

Port  $P_1$ : It was observed that in AMD  $A_2$ , iron removal percentage at  $P_1$  for 1d, 2d, 4d, 7d and 10d HRTs were 0.00%, 0.00%, 0.00%, 65.06% and 65.27% respectively and pH was found increased from 4.30 to 5.30, 5.90, 6.00, 6.90 and 6.90 respectively (Fig.4). Further ferric iron are getting converted to ferrous iron in organic substrate layer in reducing environment therefore ferric iron concentration reduced from 9.60 mg/L to 1.00 mg/L, 0.00 mg/L, 0.00 mg/L, 0.00 mg/L and 0.00 mg/L for 1d, 2d. 4d, 7d and 10d HRTs respectively. Hence same can be justified with increase in ferrous iron concentration from 86.0 0 mg/L to 94.60 mg/L, 95.60 mg/L and 95.60 mg/l for 1d, 2d, and 4d HRTs respectively up to pH level 6.00. The above phenomenon is in agreement previous researchers (Watzlaf et al., 2004). At port  $P_2$  significant iron removal was observed after 4d HRT and onwards.

Port  $P_2$ : The observed cumulative iron removal percentage for 1d,2d,4d,7d and 10d HRTs were 0.00%, 0.10%, 20.29%, 81.69% and 93.41% respectively and pH were increased to 5.80, 6.00, 6.30, 7.20 and 7.50 respectively. The significant iron removal took place at port  $P_2$  from 2d HRT and onwards. The significant reductions in ferric iron concentration were observed at port  $P_2$ . Therefore it obvious that all the ferric iron is reduced to ferrous iron in organic substrate layer and concentration of ferric iron were became zero.



Fig.4 Iron removal percentage in different ports: AMD A<sub>2</sub>

Port P<sub>3</sub>: The observations at port P<sub>3</sub> showed continue increasing trend in iron removal; with increasing HRTs. The observed cumulative removal percentage at port P<sub>3</sub> for 1d, 2d, 4d, 7d and 10d HRTs were 27.10%, 28.35%, 59.10%,100% and 100% respectively with corresponding pH increased to 6.40, 6.50, 6.80, 8.60 and 8.75 respectively. The iron removal took place at port P<sub>3</sub> after 2d HRT and onwards. Port P<sub>3</sub> is most important terminal because it discharges the treated AMD in oxidation cell and again oxic condition prevails therefore we can assess the performance of SAPS cell by analyzing the discharge from port P<sub>3</sub>. It has been observed that iron present at this stage is found in ferrous iron form. In port P<sub>3</sub>, 100% iron removal were observed for 7d and 10d HRTs whereas less removal percentage were reported for 1d, 2d and 4d HRTs therefore iron removal increases with increase in HRTs.

In AMD  $A_2$  iron removal process initiated when AMD passes through organic layer at pH level of 6.00 in anaerobic condition. The iron removal percentages were increased with increase in HRT duration along with further increase in pH as shown in Fig.4. The above results are in agreement with findings of many researchers (Jage et al., 2001). The SAPS cell effluent at port P<sub>3</sub> showed high Fe removal. Therefore the laboratory SAPS is capable for neutralization of given AMD. Highest iron removal rate was achieved for 7 days it is also observed that most of iron removed between pH range from 6.00 to 8.60 for AMD  $A_2$  as presented in Fig.5.



#### 3.2 SAPS PERFORMANCE WITH AMD B<sub>2</sub>

# 3.2.1 Oxidation reduction potential (ORP) and dissolved oxygen (DO) status in SAPS cell B

The ORP in SAPS cell B for AMD  $B_2$  showed downward trend at port  $P_3$  from 107.30 mV to - 213.50, -213.60, -312.30, -314.30 and -375.80 mV for 1d, 2d, 4d, 7d and 10 d HRTs respectively, which indicated that a strong reducing environment was prevailing inside the SAPS cell A. The negative ORP inside the SAPS is an essential condition for efficient operation of SAPS system as shown in Fig.6. The DO level in SAPS cell for AMD  $A_2$  was dropped from 5.65 to zero up to port  $P_2$  for all HRTs, which was an encouraging indicator of attainment of anaerobic conditions inside the SAPS cell. Similarly DO levels were also reported as zero at port  $P_3$  for all the HRTs as shown in Fig.7. Therefore DO and



ORP changed status is favourable for anaerobic reducing environment inside all the four SAPS cells. The total dissolved solids (T.D.S.) and sulfate were also decreasing during SAPS process which confirmed the metal removals and alkalinity generation in SAPS cell.

#### 3.2.2 Iron removal behavior

Port  $P_1$ : It was observed that in influent AMD  $B_2$ , iron removal percentage at  $P_1$  1d, 2d, 4d, 7d and 10d HRTs were 0.00%, 0.00%, 6.75%, 45.51% and 53.25% respectively and pH was found increased from 3.60 to 5.80, 5.90, 6.20, 6.60 and 6.70 respectively (Fig.7). In AMD  $B_2$  the ferric iron concentration was 30.60 mg/L and the amount of ferrous iron was 90.90 mg/ L. Initially in SAPS process up to port  $P_1$  the most of ferric iron converted to ferrous iron, these trends validate previous researcher's findings. The reductions of ferric iron were increased with respect to HRT durations. The residual ferric iron at port  $P_1$  reduced to zero for all HRTs.

Port  $P_2$ : The cumulative iron removal percentage in AMD  $B_2$  up to port  $P_2$  1d, 2d, 4d, 7d and 10d HRTs were 0.08%, 6.83%, 53.25%, 69.96% and 81.56% respectively and corresponding pH were also increased from 3.60 to 6.00, 6.20, 6.70, 7.00 and 7.20 respectively. At port  $P_2$  ferric iron reported as zero, which means all the ferric iron to convert to ferrous iron. The iron removal initiated at port  $P_2$  from 1d HRT and further increased with increasing in HRTs.

Port  $P_3$ : The cumulative iron removal percentage in AMD  $B_2$  up to port  $P_3$  for 1d, 2d, 4d, 7d and 10d HRTs were 5.19%, 45.35%, 65.19%, 100% and 100% respectively and corresponding pH were also increased from 3.60 to 6.10, 6.60,









6.90, 8.20 and 8.70 respectively as shown in Fig.9. The iron removals were reported at port  $P_3$  for all HRTs. The iron removal were initiated from 1d HRT and further increased with increase in HRTs as given in Fig.8.

#### 3.3 SAPS performance with AMD $C_2$

# 3.3.1 Oxidation reduction potential (ORP) and dissolved oxygen (DO) status in SAPS cell C

The ORP in SAPS cell C for AMD C<sub>2</sub> showed downward trend at port P<sub>3</sub> from 108.40 mV to - 211.30, -208.40, -308.30, -310.30 and -369.50 mV for 1d, 2d, 4d, 7d and 10d HRTs respectively, which indicated that a strong reducing environment was prevailing inside the SAPS cell C as shown in Fig.10. The negative ORP inside the SAPS is an essential condition for efficient operation of SAPS system. The DO level in SAPS cell for AMD C<sub>2</sub> was dropped from 6.42 to zero up to port P<sub>2</sub> for all HRTs, which was an encouraging indicator of attainment of anaerobic conditions inside the







SAPS cell. Similarly, DO levels were also reported as zero at port  $P_3$  for all the HRTs as given in Fig.11. Therefore DO and ORP changed status is favourable for anaerobic reducing environment inside all the four SAPS cells in phase II. The total dissolved solids (T.D.S) and sulfate were also decreasing during SAPS process which confirmed the metal removals and alkalinity generation in SAPS cell.

#### 3.3.2. Iron removal behaviour

Port P<sub>1</sub>: It was observed that in AMD C<sub>2</sub> iron removal percentage at port P<sub>1</sub> for 1d, 2d, 4d, 7d and 10d HRTs were 0.06%,0.12%,0.12%,35.23% and 45.27% respectively and their corresponding pH were also increased from 2.72 to 5.50, 5.70, 5.80, 6.50 and 6.60 respectively (as presented in Fig.12). Initially ferric iron concentration in AMD was 81.20 mg/L and ferrous iron concentration was 91.10 mg/L but at port P<sub>1</sub> most of the ferric iron were converted to ferrous iron, however at port P<sub>1</sub> residual ferric iron concentration reduced to 0.00 mg/L, 1.40 mg/L and 1.40 mg/L for 1d, 2d,4d and 7d HRTs respectively and ferric iron concentration reduced to 0.00 mg/L for 7d and 10d HRT all the residual ferric iron finally were converted to ferrous iron finally were converted to ferrous iron for 1d, 2d,4d and 7d HRTs respectively and ferric iron concentration reduced to 0.00 mg/L for 7d and 10d HRT all the residual ferric iron finally were converted to ferrous iron iron up to port P<sub>2</sub>.

Port P<sub>2</sub>: The cumulative iron removal percentage for AMD C<sub>2</sub> up to port P<sub>2</sub> for 1d, 2d, 4d, 7d and 10d HRTs were 27.16%, 27.57%, 39.41%, 52.41% and 69.99% respectively and their corresponding pH were increased from 2.72 to 6.40, 6.40, 6.55, 6.70 and 7.00 respectively. All the iron were in ferrous iron form at port P<sub>2</sub>, which was an essential condition of AMD



before entering the lime stone layer for efficient functioning of SAPS. There was an increasing trend in iron removal at port  $P_2$  with increasing HRTs from 1d HRT to 10d HRT.

Port  $P_3$ : The cumulative iron removal percentage in AMD  $C_2$  up to port  $P_3$  for 1d, 2d, 4d, 7d and 10d HRTs were 45.27%, 52.17%, 62.10%, 100% and 100% respectively and corresponding pH were also increased from 2.72 to 6.60, 6.75, 6.85, 8.20 and 8.60 respectively as shown in Fig.13. The complete iron removal were observed at Port  $P_3$  for 7d and 10d HRTs. Increasing trends in iron removal were observed with increase in HRT from 1d HRT to 4d HRT at SAPS cell effluent port  $P_3$ .



#### 3.4 SAPS PERFORMANCE WITH AMD D<sub>2</sub>

## 3.4.1 Oxidation reduction potential (ORP) and dissolved oxygen (DO) status in SAPS cell D

The ORP in SAPS cell A for AMD  $D_2$  showed downward trend at port  $P_3$  from 106.30 mV to - 209.40, -209.40, -307.40, -317.40 and -369.30 mV for 1d, 2d, 4d, 7d and 10 d HRTs respectively, which indicated that a strong reducing environment was prevailing inside the SAPS cell D as shown in Fig.14. The negative ORP inside the SAPS cell D as shown in Fig.14. The negative ORP inside the SAPS is an essential condition for efficient operation of SAPS system. The DO level in SAPS cell for AMD  $D_2$  was dropped from 5.65 to zero up to port  $P_2$  for all HRTs, which was an encouraging indicator of attainment of anaerobic conditions inside the SAPS cell. Similarly, DO levels were also reported as zero at port  $P_3$  for all the HRTs as given in Fig.15. Consequently DO



Fig.14 ORP variation in SAPS with AMD D<sub>2</sub>



and ORP changed status is favourable for anaerobic reducing environment inside all the four SAPS cells in phase II. The total dissolved solids (T.D.S.) and sulfate were also decreasing during SAPS process which confirmed the metal removals and alkalinity generation in SAPS cell.

#### 3.4.2 IRON REMOVAL BEHAVIOR

Port  $P_1$ : In AMD  $D_2$  initial concentration of ferric iron in influent was 105.30 mg/L and ferrous iron was 88.90 mg/l. The iron removal percentage for AMD  $D_1$  up to port  $P_1$  for HRTs 1d, 2d, 4d, 7d and 10d are 0.05%, 0.10%, 0.57%, 35.79% and 45.01% respectively and with their corresponding pH were increased from 2.50 to 5.00, 5.50, 5.60, 6.50 and 6.60 respectively. At port  $P_1$  only 8.20 mg/L ferric iron were reported as remaining because of most of ferric iron were converted to ferrous iron that is why concentration of ferrous iron were increased up to 185.90 mg/L for 1d HRT whereas 2.30 mg/L, 2.10 mg/L ferric iron were reported as residual for 2d and 4d HRTs. For 7d and 10d HRTs set up residual ferric iron were reported as nil. Significant iron removal were reported at port  $P_1$  for 7d HRT set up an onwards as shown in Fig.16.



Fig.16 Iron removal percentage in different ports: AMD D<sub>2</sub>

Port P<sub>2</sub>: Cumulative iron removal percentage for AMD D<sub>1</sub> up to port P<sub>2</sub> for HRTs 1d, 2d, 4d, 7d and 10d are 0.05%, 4.94%, 27.51%, 52.94% and 59.42% respectively with their corresponding pH increases from 2.50 to 5.80, 6.10, 6.40, 6.70 and 6.80 respectively. At Port P<sub>2</sub> only 2.10 mg/L ferric iron were reported for 1d HRT and for rest of HRT duration, no

ferric were reported, it means that all the ferric iron were converted to ferrous iron before going to next layer in SAPS. Iron removal initiated from 1d HRT and continues up to 10d HRT in increasing manner as given in Fig.17.



Port  $P_3$ : The cumulative iron removal percentage in AMD  $D_2$  up to port  $P_2$  for 1d, 2d, 4d, 7d and 10d HRTs were 5.10%%, 61.12%, 64.93%, 100% and 100% respectively and with their corresponding pH were increased from 2.50 to 6.10, 6.80, 6.90, 8.30 and 8.40 respectively. In this port of SAPS cell 100% iron removal were observed for 7d and 10d HRTs and iron removal trend is increasing with increasing the duration of HRT from 1d to 4d.

#### 4. Conclusions

A lab-scale experimental set up is made to study the metal removal, that is, iron removal in the course of the treatment of acid mine drainage. The Successive Alkalinity Producing System (SAPS) with a control ecosystem is used. After the analysis and interpretation of the collected data the main conclusions are drawn as follows:

- (1) The dissolved oxygen (DO) become zero in SAPS cell which create an anaerobic conditions that prevents armoring of limestone at the SAPS bottom, which increases the life of SAPS.
- (2) Successive Alkalinity Producing System (SAPS) has been found effective in treatment of AMD containing rich iron.
- (3) Alkalinity generation by SAPS increases with increase in iron content in influent AMD.
- (4) The net alkalinity generation increases for increase in duration of hydraulic retention time.

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