J. Mater. Environ. Sci. 7 (8) (2016) 3004-3010
ISSN : 2028-2508
CODEN: JMESCN

Design and Experimental Testing of Small-scale Acid Mine Drainage Treatment Plant

S. Nasir[1], E. Ibrahim[2], A.T. Arief[2]
1) Laboratory of Separation Engineering, Chemical Engineering Department, Faculty of Engineering, Sriwijaya University
Jl. Raya Palembang-Prabumulih Indralaya Ogan Ilir 30662, Indonesia
2) Mining Engineering Department, Faculty of Engineering, Sriwijaya University
Jl. Raya Palembang-Prabumulih Indralaya Ogan Ilir 30662, Indonesia

Received 18 Apr 2014, Revised 19 Jun 2016, Accepted 29 Jun 2016

**Corresponding author. E-mail: subriyer@unsri.ac.id. (S. Nasir); Phone/Fax: +62 711 580 303)*

Abstract

A small-scale plant consists of sand filter; ultrafiltration and reverse osmosis for treatment of Acid Mine Drainage (AMD) was designed and tested using a synthetic solution of AMD. In the current work, rice husk ash and coal fly ash were used as an adsorbent for AMD. Some parameters such as acidity (pH), Electrical Conductivity (EC), iron, manganese, and sulfate concentrations were investigated. The results showed that permeate flux and water recovery percentage obtained at operating pressure of 3.4 $kg.cm^{-2}$ as 4.06 $L.min^{-1}$ and 10.8%, respectively. It is also found that the AMD plant can increase the pH of synthetic acid mine up to 125 % and the sulfate, iron, and manganese are reduced by 98.00, 94.11, and 95.84 %, respectively. The results also showed that the AMD small scale plant was capable to produce high quality of permeate and have met the environmental standard.

Keywords: Acid mine, Reverse Osmosis, Sand Filter, Ultrafiltration

1. Introduction

The coal mining activities have increased widely in South Sumatra and other parts of Indonesia. Therefore, more acid mine water will produce in such regions and contributed to serious problem for water resources. The AMD impacts on water bodies occur not only in mining activities, but also for the abandoned mines. In the conventional method of AMD treatment, lime and limestone commonly used to neutralize the acidity. Thus, larger quantities of lime or limestone as the AMD neutralizers would have an effect on the operating cost and environmental risks as well. For instance, PT. Kaltim Prima Coal (KPC) one of the Indonesian coal mining company in 2009 spent about 2,787 tons of limestone for AMD. The amount is almost tripled compared to 2008 with the total of 1,036 tons. PTBA, another Indonesian coal mining industry located in Tanjung Enim, has been using the conventional method for AMD treatment, namely the addition of lime into the AMD (pH 3-4) in the neutralization ponds prior releasing water into the river. Lime and limestones are an inexpensive reagent for AMD, and a lower volume of sludge is produced as a side product of its application. However, limestone is not as effective as AMD neutralizer when the AMD has a high concentration of ferrous iron and the use of limestone is more complex than lime in the process. Limestone is more difficult to raise the pH 6 and above due to carbon dioxide formation during acid neutralization [1].

The AMD treatment can be classified into active and passive treatments. Active treatment refers to the monitoring process based on external sources of energy using the available infrastructure and system engineering such as aeration, neutralization, chemical precipitation, membrane processes, ion exchange and biological sulfate removal. The passive treatment is a process with minimum human effort in both operations and maintenance. It takes an advantage of the natural chemical and biological processes that ameliorate contaminated water conditions [2-5]. Due to the nature and characteristics of AMD, it is cannot be solved in the short term application or by single intervention. Therefore, it needs a more comprehensive, process integration and sustainable handling of AMD problems [6].

The AMD has a low acidity and high concentration of soluble metal cations and sulfate ions. The presence of sulfate ions originates from coal sulfur contents. The sulfur in coal sediment was found in the form of pyrite

(FeS_2), marcasite (FeS_2), pyrrhotite (FexSx), chalcosite (CuS_2), covelite (CuS), chalcopyrite ($CuFeS_2$), molybdenite (MoS_2), milerite (NiS), galena (PbS), sphalerite (ZnS), and arsenopyrite (FeAsS) [7,8].

Ultrafiltration application is often found in electrocoating paint, biodiesel purification, fruit juice clarification, separation of oil and water emulsions, water purification, and enzyme production [7-13]. The UF and RO combine is sufficient to remove suspended solids, bacteria, and colloid in copper mine wastewater. Therefore, it can be used to prepare the RO feed water with low turbidity and low SDI [14]. Moreover, secondary effluent treatment system using RO has become the main focus by experts in the last decade. In developed countries the reuse of excess water and wastewater are becoming popular to meet the needs of water for agricultural and industrial purposes or for soil-water enrichment [15]. Therefore, the reuse of excess water and wastewater as RO feed water also has several advantages such as reducing the amount of water extracted from the ground and avoid the need of construction and development of new water supply [16]. The treated secondary effluent using membranes proved to be used as non-potable water systems such as cooling towers or process water [17]. However, the recycling process of secondary effluent uses RO systems needs a more complex pretreatment because of the wastewater characteristics itself. Another study of the post-mining of brown coal waste treatment using RO spiral wound membranes shows that ammonia nitrogen removal efficiency was found as 94% for monovalent ions and 99 % for divalent ions respectively with TDS removal about 99.3% [18]. Although, there is no effect of AMD anions on the permeate flux when the wastewater containing Cu ions were treated using the combination of Nanofiltration (NF) and RO [19].

An attempt to neutralize the AMD has been carried out by researchers such as using lignite as an adsorbent [20], fly ash, natural clinker and synthetic zeolites [21], electrochemical [22], upflow anaerobic sludge blanket reactor (UASB) [23], using sawdust and cardboard [24], and the relatively new method of using wood ash [25], metallurgical slags [26], and steel slag [27]. The RO application has been well established for several decades, but the direct application of RO systems for AMD treatment is extremely high risk of failure because the RO membrane properties are more susceptible to the low pH of AMD. Therefore, it requires a pretreatment system of the feed water as specified by the membrane. The use of plants to reduce the metal ions in contaminated water has also been studied by researchers [28-31]. It was found that phytoremediation can decrease heavy metals and sulfate and AMD contamination can be reduced. However, such methods require a long process time and large area.

In the current research, we proposed the methods for AMD treatment using sand filtration (SF) equipped by rice husk-ash (RHA) and coal fly-ash (CFA) adsorbent columns followed by Ultrafiltration and Reverse Osmosis.

2. Materials and methods

2.1. Samples

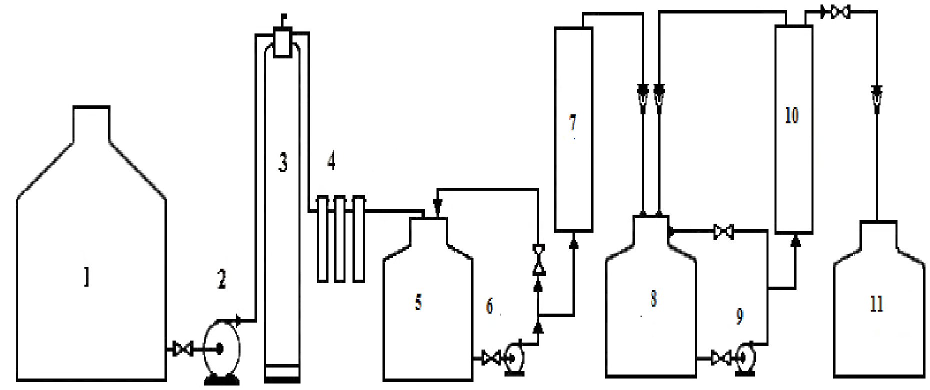
Two types of samples were used in the study. *First*, AMD (pH 3.93) was collected from the Udongan Treatment Pond coal mining area in Tanjung Enim, South Sumatra Indonesia. *Second*, the synthetic solution of AMD samples (pH 2.4; 3.74; 3.93) was prepared by dissolving $FeSO_4\ 7H_2O$, $Al_2(SO_4)_3\ 18H_2O$ and $MnSO_4\ H_2O$ into clean water. Sulfuric acid was added into solution for pH adjustment. Samples were placed in a polyethylene tank with a capacity of 550 L. Two adsorbents were used in this work, which were RHA and CFA. The adsorbents were used without pretreatment except for grinding into 20 mesh.

2.2. Equipments

There were three major equipments used in the current work which is SF, UF, and RO. *First*, the rapid SF (type FRP EL88JO) consists of a fiber tube with dimensions of 140 cm height, 25 cm of width, and 0.2 cm of thickness. The tube filled with 30% wt of activated carbon and 70% wt of silica-sand. Adsorption column (51 cm of length and 7.5 cm of diameter) filled by RHA and CFA, respectively. *Second*, the UF system (type HM90UF, MWCO 50,000–150,000 Daltons, 0.3404 m^2 of membrane area) consists of a hollow fiber membrane placed in a stainless - steel housing with dimension: 184 cm of height, 12 cm of diameter and 0.2 cm of thickness. *Third*, Reverse Osmosis unit consists of a stainless booster pump and CSM-RO membrane (RF 4021–TE, 0.1808 m^2 of membrane area, South Korea).

2.3. Methods

Standard equipments for the permeate and retentate quality analysis used in this study were pH meter, Electrical Conductivity Meter, Analytical Balance, and UV-200 RS Spectrophotometer. The parameters tested were pH, electrical conductivity, and concentration of iron, manganese, and sulfate ions. The variables investigated in this study were feed flow rate, operating pressure, and operating time. Feed flow rate for the rapid SF was constant (10 $L.min^{-1}$). The operating pressure of UF was set at 2.5; 3. 5 and 4.5 $kg.cm^{-2}$ while the operating time was set for one hour. Permeate from RO was collected by using a measuring cup with a volume of 1000 mL and analyzed in interval of 15 minutes. Manganese, iron, and sulfate concentration were measured using UV-Vis type 200RS spectrophotometer at wavelength of 425, 540 and 420 nm respectively. Schematic of AMD small-scale treatment plant designed is shown in Fig. 1.



1. Feed tank 2,6,9 Centrifugal tank 3. Sand filter 4. Adsorbent column 5. UF feed tank 7. UF module 8. RO feed tank 10. RO Module 11. Permeate Tank

Figure 1: Schematic of Small-scale AMD treatment plant

The following formula was used to calculate the removal efficiency (RE) of Fe, Mn and Sulfate ions of Sand Filter, Ultrafiltration and Reverse Osmosis.

$$RE\ (\%) = (A - B)/A \times 100 \qquad (1)$$

Where: A is the initial concentration of ions (ppm), B is the final concentration of ions in the permeate.
Total of average removal efficiency (RE_{total}) for the overall system is calculated using the formula.

$$(RE)_{total}\ (\%) = (RE_{SF}) + RE_{UF} + RE_{RO})/3 \qquad (2)$$

Water recovery (WR) is calculated with the following formula:

$$WR\ (\%) = PF/(PF+RF) \times 100 \qquad (3)$$

Where PF = Permeate Flowrate ($L.h^{-1}$), CF = Retentate Flowrate ($L.h^{-1}$)

3. Results and discussion

3.1. AMD Analysis

The parameters tested were pH, TSS, TDS, EC, turbidity, and concentrations of Fe, Mn, and sulfate ions. AMD samples analysis is shown in Table 1.

Table 1: AMD Analysis

No.	Parameter	Unit	Real AMD	Synthetic Solution AMD	Indonesian Standard
1.	Turbidity	NTU	35.3	-	-
2.	TDS	mg/L	1650	-	-
3.	TSS	mg/L	14.2	-	400
4.	pH	-	3.93	3.93 ± 0.1	6-9
5.	Iron	mg/L	0.81	0.81± 0.05	7
6.	Manganese	mg/L	10.35	10.35±0.05	4
7.	Sulfate	mg/L	1340.9	1340.9± 0.1	-
8.	Aluminium	mg/L	1.603	-	-

The real AMD and a synthetic solution AMD samples analysis are shown in Table 1. AMD samples show that the level of TDS, pH, iron, manganese, aluminum, and sulfate ions are quite high. However, turbidity, TDS, aluminum, and sulfate is not listed or regulated by Indonesian National Standard of coal wastewater. AMD samples also shown that TSS is extremely lower than permitted range in the Indonesian standard therefore it is not considered in the current work. Similar to TSS, turbidity of AMD samples are not investigated in the present study because their effect to the membrane fouling possibility were low. Another reason is the sand filter as a pretreatment will reduce the amount of TSS of AMD samples. Due to a high volume of AMD (550 L for each run) to conduct the experiments, synthetic solution of AMD with pH 3.93 were prepared and tested for all the experiments.

3.2. Permeate Quality

Table 2 shows the SF filtrate analysis at a different feed flow rate. It can be seen that the SF filtrate has stabilized pH after one hour of contact time. The SF is capable to increase the pH of AMD from 3.93 to 5.75. This is because the porous characteristics of activated carbon in the SF which adsorb the metal ion in the sample. Meanwhile, silica sand was effective to reduce the TDS and TSS of AMD and substantially improved the turbidity of AMD. However, increases of feed flowrate to SF will slightly increase the acidity of filtrate. Nevertheless, filtrate acidity still below the standard permit. This is suggested that the higher feed flow of samples will reduce the contact between feed solution with activated carbon and silica sand in the SF.

Table 2: Analysis of SF filtrate

Filtrate flowrate of Sand Filter (L.min^{-1})	3.0		3.3	
Time (Min.)	pH (-)	EC (µs.cm^{-1})	pH (-)	EC (µs.cm^{-1})
15	5.5	1504	5.6	1936
30	6.0	1847	5.8	1910
45	6.0	1936	5.8	1905
60	6.0	1994	5.8	1902
Average	5.86	1820.25	5.75	1913.25

Table 3 illustrates that the pH of UF permeates is in the range of 6.0 to 6.2 when RHA and CFA used as adsorbents. Further treatment of UF permeates using RO shown that RO able to increase the pH of permeates from 5.8 up to 6.5 for RHA and 6.8 for CFA. It means that RO can effectively reject iron, manganese, and sulfate ions from UF feed solution as shown in Fig. 2. Table 3 also illustrates the pH of AMD after a filtration using SF, UF and RO with RHA as an adsorbent. As suggested an increase of contact time will increase the pH and decreases EC of permeates. It is shown that the permeates from UF have pH and EC are ranging from 6.0 to 6.2 and 1818 to 1994 µS.cm^{-1}, respectively. It means that UF is able to provide permeates which met the environmental standard. Moreover, further treatment of UF permeates using RO and CFA as an adsorbent which can increase the pH of synthetic acid mine up to 6.8 after one hour of contact time.

Table 3: Analysis of Permeate and Retentate

RHA					CFA			
Ultrafiltration								
Time (Min.)	Permeate (L.min^{-1})	Retentate (L.min^{-1})	pH (-)	EC (µs.cm^{-1})	Permeate (L.min^{-1})	Retentate (L.min^{-1})	pH (-)	EC (µs.cm^{-1})
15	3.5	7.5	6.0	1516	2.5	9.8	6.0	1931
30	3.5	7.5	6.0	1707	2.5	9.8	6.1	1913
45	3.5	7.5	6.0	1797	2.5	9.0	6.2	1911
60	3.5	7.5	6.2	1818	2.5	9.0	6.2	1907
Reverse Osmosis								
15	0.04	6.9	6.0	49.2	0.28	6.25	6.5	10.25
30	0.05	6.9	6.5	45.1	0.27	6.25	6.7	8.69
45	0.05	6.9	6.5	42.0	0.25	6.25	6.7	12.29
60	0.05	6.9	6.5	40.6	0.26	6.25	6.8	15.68

Fig. 2 shows that RO has a high removal efficiency of iron, manganese and sulfate ions compared to those of SF and UF. The UF membrane is not effective to reduce divalent ion in feed water, however, UF can be used as pretreatment for RO. It can be seen from Fig. 2 that the average removal efficiency of iron, manganese and sulfate ions as 94.11, 95.84, and 98.35% , respectively when the CFA was used as a sorbent. CFA has a spherical shape particle with metal oxide as a major compound [21]. This suggested that CFA has a wide range of pore distribution which adsorb the heavy metal ions on the adsorbent surface. Calcium oxide in CFA reacts with sulfate ions in aqueous solution and form calcium sulfate. More increase of CaO concentration in CFA will decrease the sulfate ion in acid mine thus neutralize the acidity of AMD.

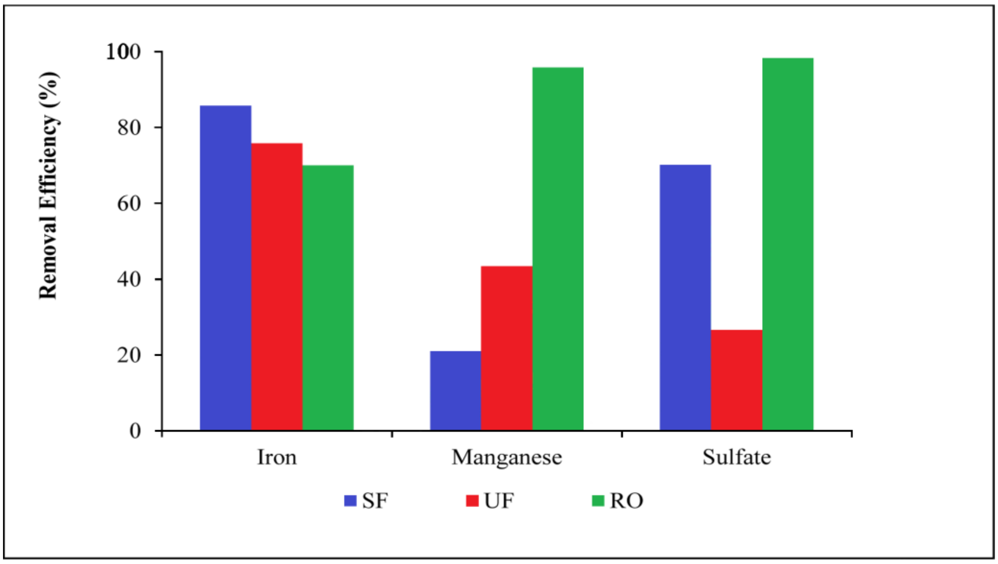


Figure 2: Removal efficiency of Iron, Manganese and Sulfate ions

Fig. 3 represents the pH and EC removal efficiency percentage. As seen in Fig 2. RHA and CFA are good sorbents for AMD. However, CFA more effective for EC and pH removal. Typically, RHA contains almost 90% of silica oxide, and efective to reduce the heavy metal such as iron (Fe^{2+}, Fe^{3+}), Zn^{2+} and Cu^{2+} [32]. Otherwise, metal oxide concentration such as CaO is greater in CFA compared to those of RHA. CFA in this investigation is categorized as Class C of fly-ash which contains 30-40% of CaO [33]. Therefore, CaO in CFA has similar characteristic with lime and limestone and capable to neutralize the acidity of AMD.

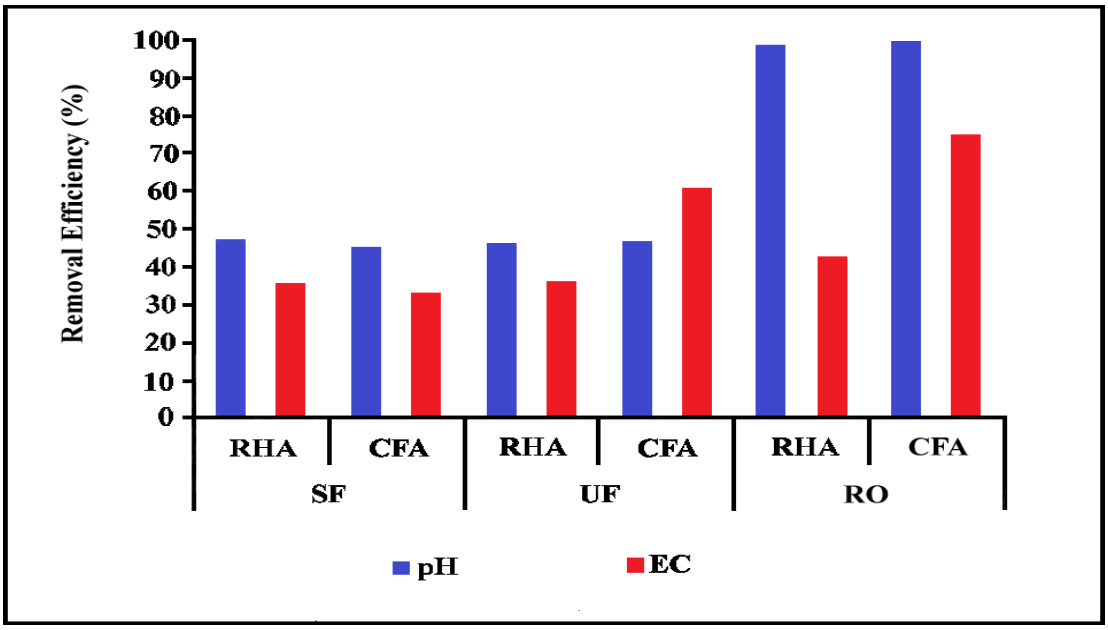


Figure 3: Removal efficiency of EC and pH

3.3. Water Recovery Percentage

Fig. 4 shows the water recovery (WR) percentage of UF and RO at different operating time. Water recovery both UF and RO will increase with operating time. The water recovery percentage of UF are higher compared to RO. The low water recovery percentage of RO obtained because of low applied operating pressure (about 3.4 kg.cm^{-2}) and small membrane area of RO. It can be concluded that successful of SF as pretreatment affected the quality of feed water for UF and RO thus reduce the fouling propensity and increase the membrane life.

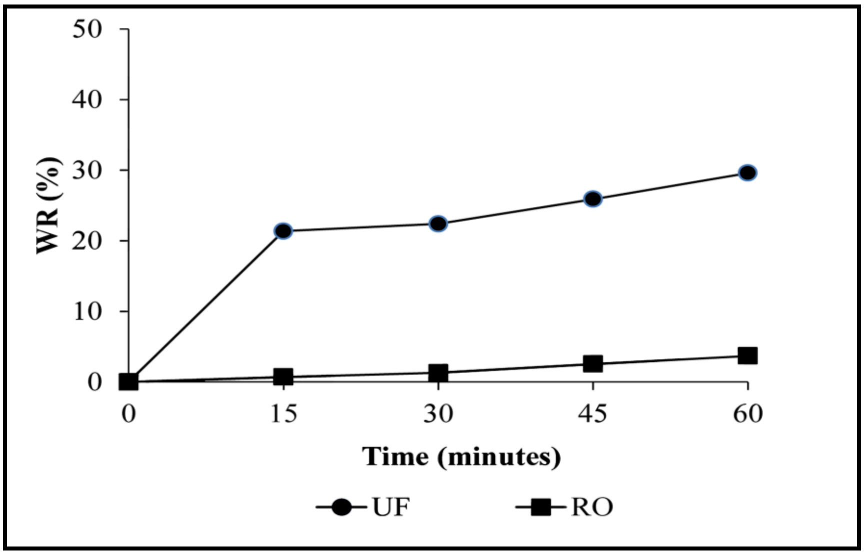


Figure 4: Water recovery percentage

Conclusion

The use of rice husk ash and coal fly ash as an adsorbent for the AMD pretreatment combining with ultrafiltration and reverse osmosis was able to increase the final pH permeate of RO in the range of 6.0 to 6.8. The application of the small scale of AMD treatment plant designed in this work was able to reduce iron, manganese, and sulfate concentrations up to 94.11, 95.84, and 98.00%, respectively. It also concluded that the RO permeates are safely to discharge into the environment or can be used as process water.

Acknowledgements-Author thanks Directorate of Research and Community Service, Ministry of Research, Technology and Higher Education, Republic of Indonesia through Penelitian Unggulan Perguruan Tinggi Negeri UNSRI, for supporting this work.

References

1. Akcil, A., Koldas, S., *J. Clean. Prod.*, 14 (2006) 1139-1145.
2. Watzlaf, G.R., Schroeder, K.T., Kleinmann, R.L.P., Kairies, C.L., Nairn, R.W., The Passive Treatment of Coal Mine Drainage, DOE/NETL-2004/1202, U.S. Department of Energy, National Energy Technology Laboratory, Pittsburgh, PA, (2004)
3. Younger, P.L., Banwart, S.A., Hedin, R.S., Mine Water: Hydrology, Pollution, Remediation, The Netherlands: Kluwer Academic Press, (2002).
4. Costello, C. Acid Mine Drainage : Innovative Treatment Technologies, U.S. Environmental Protection Agency Office of Solid Waste and Emergency Response Technology Innovation Office, Washington DC, (2003).
5. Skousen, J., Rose, A., Geidel, G., Foreman, J., Evans, R., Hellier, W., Handbook of Technologies for Avoidance and Remediation of Acid Mine Drainage, The National Mine Land Reclamation Centre, Morgantown, West Virginia, (1998).
6. Johnson, D. B., Hallberg, K.B., *Sci. Tot. Environ.*, 338 (2005) 3–14.
7. Simate, G.S., Ndlovu, S., *J. Env. Chem. Eng.*, 2 (2014) 1785-1803.
8. Yeh, M.H., *Desalination*, 145 (2002) 153–157.
9. Vela, M.C.V., Blanco, S.A., García, J.L., Gozálvez-Zafrilla, J.M., Rodríguez. E.B., *Desalination*, 204 (2007) 328–334.

10. Gomes, M.C.C., Arroyo, P.A., Pereira, N.C., *J. of. Memb. Sci.*, 496 (2015) 242–249.
11. Zhang, J.C., Wang, Y.H., Song, L.F., Hu, J.Y., Ong, S.L., Ng, W.J., Lee, L.Y., *Desalination*, 174 (2005) 247–256.
12. Langenbach, K., Kuschk, P.,Horn, H., Kästner. M., Water. Res., 44 (2010) 159-166.
13. Echavarría, AP., Falguera, V., Torras., C, Berdún., C, Pagána., J, Ibarz., *LWT - Food Sci.Technol.*, 46 (2012) 189–195.
14. Xu, J., Ruan, L.G., Wang, X., Jiang, Y.Y., Gao, L.X., Gao, J.C., *Sep. Purif. Technol*., 85 (2012) 45–53
15. Cséfalvay, E., Pauer, V., Mizsey, P., *Desalination*, 240 (2009) 132-142.
16. Shao, E., Wei, J., Yo, A., Levy, R., Application of Ultrafiltration and Reverse Osmosis for Mine Waste Water *in IWWA Proceeding of Reuse Water in Mining*, Perth, Western Australia, (2009).
17. Toze, S., *Agr. Water. Manage*., 80 (2006) 147-159.
18. Petala, M., Tsiridis, V., Samaras, P., Zouboulis, A., Sakellaropoulos, G.P., *Desalination*, 195 (2006) 109-118.
19. Qin, J.-J., Htun Oo, M., Nyunt Wai, M., Lee, H., Hong, S.P., Kim, J.E., Xing, Y., Zhanga, M., *Desalination*, 171 (2005) 299-305.
20. Mohan, D., Chander, S., *J. Hazard. Mat*., 137 (2006) 1545-1553.
21. Ríos, C.A, Williams, C.D., Roberts, C.L., *J. Hazard. Mat*., 156 (2008) 23–35.
22. Buzzi, D.C., Viegas, L.S., Rodrigues, M.A.S., Bernardes, A.M., Tenório, J.A.S., *Min. Eng*., 40 (2013) 82–89.
23. Rodriguez, R.P., Oliveira, G.H.D., Raimundi, I.M., Zaiat, M., *Int. J. Biodeter. & Biodeg*., 74 (2012) 48–53.
24. Markovic´, R., Stevanovic´, J., Stevanovic´, Z., Bugarin, M., Nedeljkovic´, D., Grujic´, A., Stajic´-Trošic, J., *Mat. Transact*., 52 (2011) 1849-1852.
25. Name, T., Sheridan, C., *Min. Eng*., 64 (2014) 15-22.
26. Heviánková, S., Bestová, I., Kyncl, M., *Min.Eng*., 56 (2014) 109-111.
27. Goezt, E.R, Riefler, R.G., *Chem. Eng. J*., 240 (2014) 579-588.
28. Abreu, M.M., Tavares, M.T, Batista, M., *J. Geochem. Expl*., 96 (2008) 210-222.
29. Bharti, S., Banerjee, T.K., *Ecotox. & Environ. Safe*., 81 (2012) 36-42.
30. Bech, J., Duran, P., Roca, N., Poma, W., Sánchez, I., Pérez, L.R., Boluda, R., Barceló, J., Poschenrieder, C., *J. Geochem. Explor*., 123 (2012) 109-113.
31. Shim, D., Kim, S., Choi, Y.I., Song, W.Y., Park, J., Youk, E.S., Jeong, S.C., Martinoia, E., Noh, E.W., Lee, Y., *Chemosphere*, 90 (2013) 1478-1486.
32. Mohamed, R.M., Mkhalid, I.A., Barakat, M.A., *Arab. J. Chem*., 8 (2015) 48-53.
33. Ahmaruzzaman, M., *Prog. Energy Combust. Sci*., 36 (2010) 327-363.

(2016) ; http://www.jmaterenvironsci.com